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COAL GASIFICATION/GAS CLEANUP TEST FACILITY: VOLUME IV. A MATHEMATICAL MODEL OF THE PACKED COLUMN ACID GAS ABSORBER

by

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16. ABSTRACT **The report describes a mathematical model for adiabatic operation of a packed-column absorber designed to remove acid gases from coal gasification crude product gas. It also gives results of experiments with a small pilot-scale coal gasification/gas cleaning facility designed to test the model. The model predictions compared well with the actual absorber liquid temperature profile and outlet gas composition. The model is useful for the evaluation of the effect of changes in process variables on absorber column performance and (hence) for column design.**

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Mathematical Models		Stationary Sources	12A	
Gases	Absorbers (Equipment)	Gas Absorbers	07D	13I
Test Facilities	Columns (Process Engineering)		14B	
Coal Gasification			13H	
Gas Scrubbing			07A	

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ABSTRACT

Gasification of coal produces a product that contains the acidic compounds carbon dioxide and hydrogen sulfide, as well as the primary gases carbon monoxide and hydrogen. Data show that significant amounts of other constituents such as hydrocarbons, mercaptans, and carbonyl sulfide are also produced. The removal of acid gases and other sulfur compounds is accomplished by contacting the gaseous product with a solvent in an acid gas removal system; an absorber is invariably the major unit in the system.

The pilot plant facility used to investigate the environmental factors related to gasification of coal was described in an earlier report (Ferrell et al, 1980). In this facility, the packed column absorber used in the acid gas removal system was operated adiabatically. Because of the critical role of this unit in the overall operation, a mathematical model was developed for its description. The model was based on an earlier work by Treybal (1968) and was programmed for use in the analysis of system performance. The model was developed for use in two complementary systems: those in which the carrier gas may be considered insoluble and those in which all of the system components can be absorbed or volatilized. These programs are SIMPAK and MCOMP, respectively.

Experimental data were obtained that allowed the model predictions to be tested. A gas mixture consisting of nitrogen and carbon dioxide was contacted with refrigerated methanol to evaluate SIMPAK. Alternatively, the product gas from the gasification of char or coal was contacted with refrigerated methanol in the absorber to provide data that could be compared with predictions from MCOMP. In all cases, model predictions were excellent. These results indicate that the model may be used in the design of packed adiabatic absorbers or in the analysis of experimental data from an existing facility.

As an additional feature, the use of these programs required that physical property packages be developed for constituents found in the product from a coal gasifier. These packages should be useful in a variety of applications involving such materials.

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INTRODUCTION

As a part of a continuing research program on the environmental aspects of fuel conversion, the U. S. Environmental Protection Agency has sponsored a research project on coal gasification at North Carolina State University in the Department of Chemical Engineering. The facility used for this research is a small coal gasification-gas cleaning pilot plant. The overall objective of the project is to characterize the gaseous and condensed phase emissions from the gasification-gas cleaning process, and to determine how emission rates of various pollutants depend on adjustable process parameters.

The plant was described in detail in Volume I of this report series (EPA-600/7-80-046a), and consists of a fluidized bed reactor, a cyclone and venturi scrubber for particulates, condensables, and solubles removal, and absorption and stripping columns for acid gas removal and solvent regeneration. The plant has a nominal capacity of 23 kilograms of coal feed per hour (50 lb/hr) for steady state operation. A schematic diagram of the gasifier, the acid gas removal system (AGRS), and other major components is shown in Figure 1a.

In an initial series of runs on the gasifier, a pretreated Western Kentucky No. 11 coal was gasified with steam and oxygen. The results of this work and a detailed listing of the project objectives are given by Ferrell et al., Vol II, (1981), and were presented at the EPA Symposium on Environmental Aspects of Fuel Conversion Technology V, held in St. Louis, Mo., September, 1980.

Volume III of this report series presented a detailed discussion of a series of runs made with a New Mexico subbituminous coal. As a part of the volume III report, a summary of a mathematical model of the operation of the packed tower absorber of the acid gas removal system was presented.

It is the purpose of this report to present a detailed description of the absorber model, its development, and a comparison of predictions made using the model with experimental data from the pilot plant.

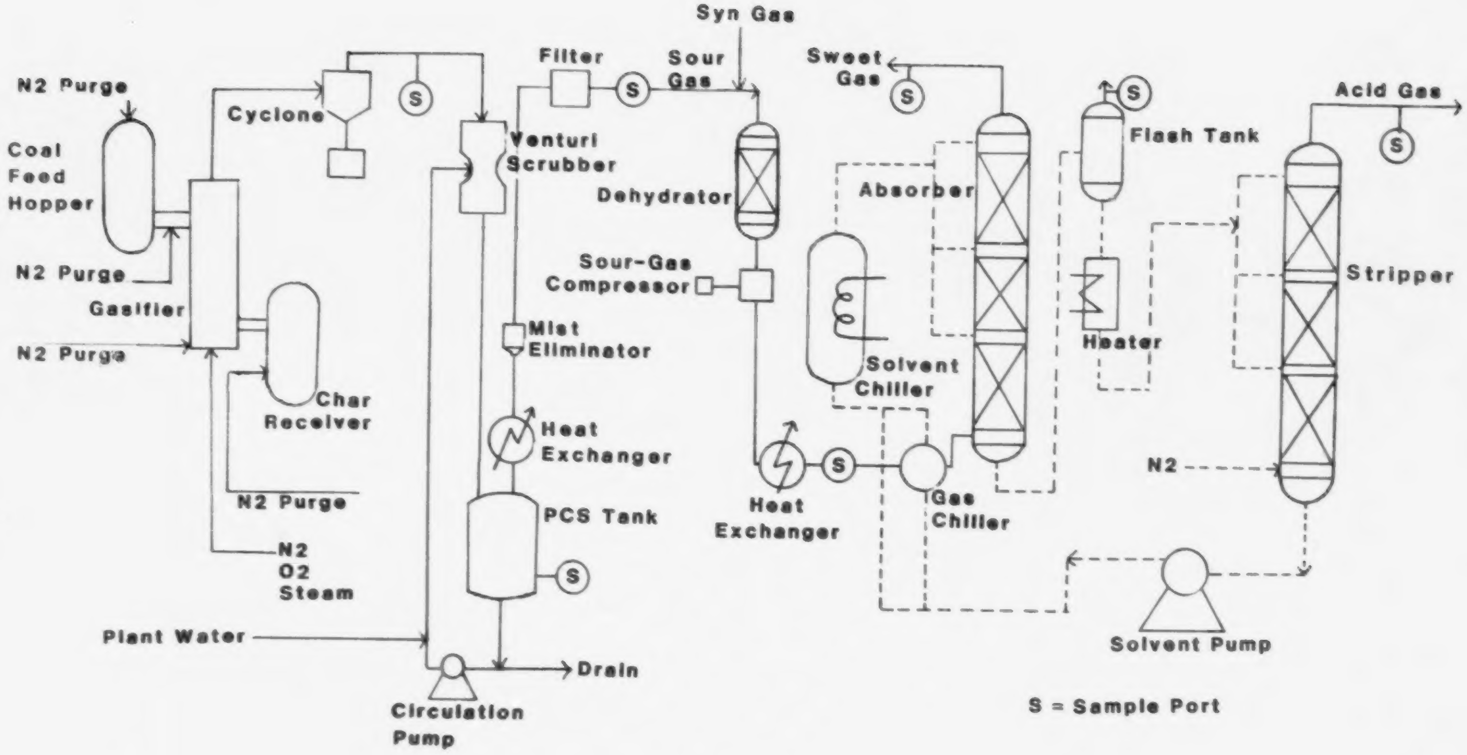

Syn Gas
N2 Purge
Coal
Feed
Hopper
Cyclone
S
Filter
S
Sour
Gas
Sweet
Gas
S
S
Acid Gas
S
Venturi
Scrubber
Dehydrator
Absorber
Flash Tank
N2 Purge
Gasifier
N2 Purge
Sour-Gas
Compressor
Stripper
Mist
Eliminator
Heater
Solvent
Chiller
Char
Receiver
Heat
Exchanger
Heat
Exchanger
S
Gas
Chiller
N2
N2 Purge
PCS Tank
N2
O2
Steam
S
Plant Water
Drain
Circulation
Pump
Solvent Pump
S = Sample Port

Figure 1a

Pilot Plant Facility

NON-ISOTHERMAL GAS ABSORPTION

Mathematical description of non-isothermal gas absorption in a packed column must consider complex fluid mechanics, heat transfer, molecular diffusion, phase equilibria and, in some cases, chemical reaction kinetics. In the case of physical absorption, the process can be viewed as "a liquid flowing downward through the packing in the form of a film, for which the thickness, the rate of movement and also the deviation of the direction of motion from the vertical are different at different points in the packing" (Danckwerts, 1967). At any point in the packing where there is a gas liquid interface, the gas can dissolve in the liquid. The amount of gas absorbed depends upon the film thickness, the diffusion potential, the solubility of the gas in the liquid, and several other factors. A physical model that will represent the gas absorption process in a packed column must account for the complicated sequence of events taking place.

TWO-RESISTANCE THEORY

The "two-film theory", as first proposed by Whitman and Lewis (1923, 1924), provides the basis for most models used today. The theory describes the absorption ot a solute gas component in the context of a series of resistances. The solute must pass through a gas-phase resistance and then through a liquid-phase resistance in its attempt to achieve equilibrium. At the interface, the gas and liquid phases are assumed to be in equilibrium.

The relative importance of the gas-phase and liquid-phase resistances depends on the solubility of the gas in the liquid and the transport parameters of the solute in each phase. For very soluble gases, the gas-phase resistance controls while for relatively insoluble gases, the liquid-phase resistance controls. For cases of intermediate solubility, both resistances must be considered. Situations involving gases of intermediate solubility are commonly encountered, although the predominance of one resistance is often assumed for the sake of simplicity.

NON-ISOTHERMAL CONSIDERATIONS

For both tray towers and packed columns, calculational techniques exist that are sufficiently accurate for situations in which gas absorption occurs isothermally, involves transfer of only one solute, results in negligible changes in gas and liquid flow rates through a column, and involves gas and liquid phases that can be treated ideally. However, where these conditions are not met, more complex approaches must be taken.

While departure from any of the above conditions introduces added complexity, large thermal effects are often the most difficult to handle in modeling absorption processes. In these cases, the release of latent heat and heat of solution raises the interfacial temperature above that of the bulk liquid. As a result physical properties, mass transfer coefficients and equilibrium compositions change (Treybal, 1980). Other factors that accompany non-isothermal absorption include volume changes (Verma and DeLancey, 1975) and diffusion (DeLancey and Chiang, 1968, 1972).

Bourne and Coggan (1969) point out that thermal effects associated with gas absorption are comprised of energy liberated by the dissolving gas, sensible heat transfer between the two phases, and transport of latent heat from evaporation or condensation of solvent. In many cases, heat transfer from the fluid to the column walls or to cooling coils is also a factor.

The simplest way to approach a non-isothermal packed or tray tower absorber calculation is to ignore these thermal processes. The column can be assumed to operate at the temperature of the entering or exiting liquid, or the average of the two, and a McCabe-Thiele diagram can be constructed to describe the system. The accuracy of this approach varies, but problems that may arise can be seen in Figure 1. The equilibrium lines calculated for the temperature of the entering and exiting liquid stream, as well as the average of the two, are compared to the rigorously calculated equilibrium line for an experimental run discussed later in this report. None of the three simplifying assumptions accurately reflects the situation within the column, and use of any of them in the calculation would lead to error. Instead, a more complex approach must be used. Nomenclature used in the development of the model is given at the end of this report.

DESIGN OF PACKED TOWERS

Sherwood and Pigford (1952) and Sherwood et al. (1975) describe a design procedure for adiabatic absorption of a single solute in a packed tower. Their procedure neglects liquid phase resistances to mass and heat transfer, and changes in liquid and gas flow rates through the tower are ignored. Dodge and Fellinger and Stuke (1941, 1953) have shown experimentally that neglecting the liquid phase resistance to mass transfer can lead to error, even in the case of very soluble gases; liquid phase resistance to heat transfer is also important. The work of Sherwood and Pigford (1952) represents the first attempt to incorporate nonisothermal effects into a generalized design procedure.

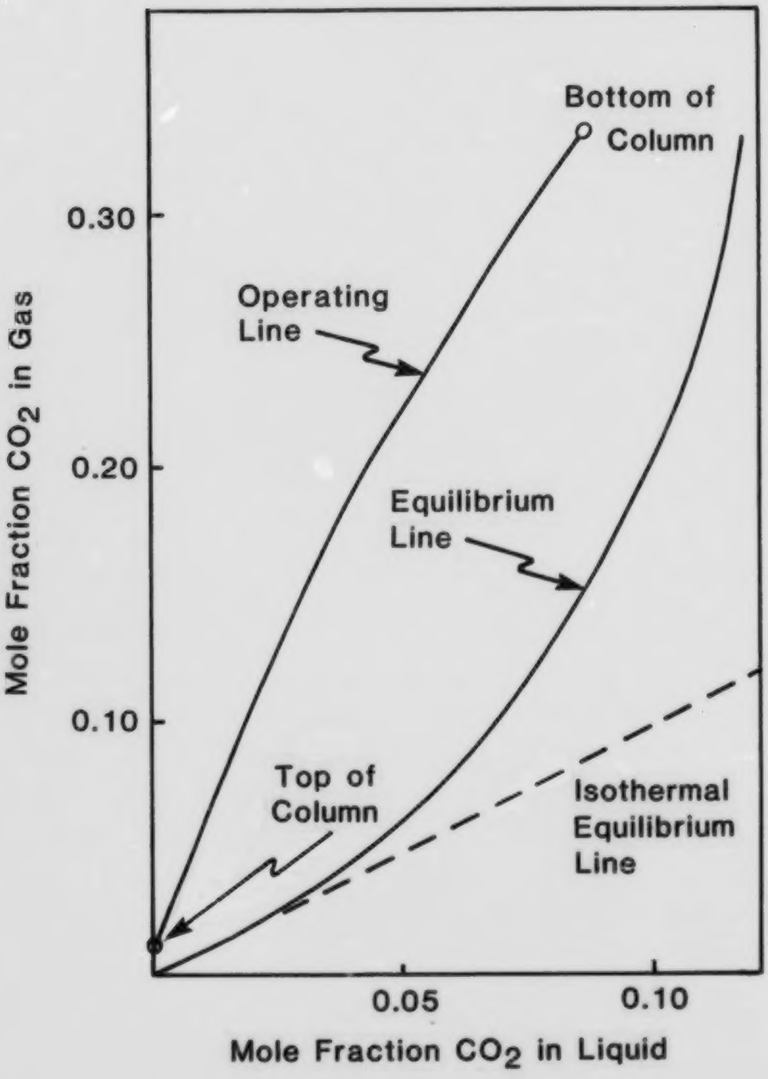
Several workers suggested techniques based on the method of Sherwood and Pigford (1952). Mizushima et al (1959) have adapted the simple adiabatic model of Sherwood and Pigford (1952) so that it can be solved graphically.

Figure 1

McCabe-Thiele Diagram

Adiabatic Absorption of CO_2 in Methanol

Computer Simulation of Run AM-32



By using a modified enthalpy expression as the driving force, they presented a method by which the total interfacial area could be determined. Hirai (1968) extended the simple adiabatic model to a system where the controlling mass transfer resistance was in the liquid phase, neglecting gas phase resistances to heat and mass transfer. These workers also discuss a case which includes the simultaneous absorption of a second solute, albeit only for low mass transfer rates of the second solute. While good agreement between experimental and predicted concentration profiles was achieved, poor agreement between experimental and predicted temperature profiles resulted.

Holland and co-workers (1970, 1969) used a tray tower analogy for packed tower calculations. They note that overall mass transfer coefficients are inappropriate in describing packed tower absorption; the inaccuracy introduced by ignoring the resistance of one phase can be substantial. Instead, an extension of Peters′ (1922) HETP (Height Equivalent to a Theoretical Plate) approach was used. Their model calls for the packed tower to be divided into various mass transfer sections that are analogous to trays in a tray tower. The mass transfer taking place in each section can be expressed in terms of a vaporization efficiency (Holland, 1970) which must be determined experimentally. Vaporization efficiency accounts for the difference between the liquid and gas leaving an ideal tray or ideal mass transfer section and that leaving a real tray or real mass transfer section. This method has been used successfully to simulate packed absorber and packed distillation column behavior during both steady and unsteady state operations for multicomponent systems. Its main disadvantage is that it requires experimental data to determine the set of vaporization efficiencies needed in the calculation.

To avoid the convergence problems with the iterative procedure used to solve the highly coupled and nonlinear system of equations for non-isothermal absorption, von Stockar and Wilke (1977) applied the relaxation technique of Bourne et al (1974) to packed tower operation. The technique simulates the process start-up and convergence is, ultimately, guaranteed. Comparisons of their modeling results to those from the simple adiabatic model of Sherwood and Pigford (1952) shows the importance of incorporating all heat effects into the design calculation. Use of their model for multicomponent cases has not been reported.

von Stockar and Wilke (1977) have also reported the use of a short-cut design method for non-isothermal absorption in packed towers. They use a semi-empirical correlation first to predict the maximum temperature in the column and then to predict the entire temperature profile. This information is combined with a transfer unit approach to calculate packing height required to accomplish a specified separation. Their approach allows non linear equilibrium relationships, and in cases where solvent volatility is negligible, further simplifications are possible. They showed that the short-cut method is superior to the isothermal and simple adiabatic models for 90 hypothetical design cases using the rigorously calculated height of packing as the point of comparison.

A calculational technique which treats a packed tower as a true differential device is described by Treybal (1969) for the absorption of a single solute under adiabatic conditions. This approach is based on the work of Olander (1960, 1961) who dealt with a problem involving no mass transfer in the liquid. The method considers the mass and heat transfer resistances of both gas and liquid phases and assumes that axial dispersion is negligible. Evaporation and condensation of the solvent is included in the model and the equivalence of interfacial areas for mass and heat transfer is assumed. Treybal (1969) presented a sample calculation for the air-water-ammonia system and suggested the extension of the technique to handle multicomponent cases. Using this approach, Raal and Khurana (1973, 1970) provided experimental verification of model predictions for the air-water-ammonia system. Excellent agreement between the model and experimental data was found once a modified correlation for the interfacial area was adopted.

Feintuch and Treybal (1978, 1973) improved the earlier models to include the calculation of the minimum solvent requirements; they developed a computer algorithm for describing adiabatic absorption for both single solute and multicomponent cases. The data of McDaniel (1970), which had been obtained from a fourteen-component non-isothermal absorption column, was used to test the algorithm; discrepancies in the column material and energy balances prevented conclusive verification of the model. A short-cut calculation for complex multicomponent adiabatic systems, based on the absorption factor method of Edmister (1943) was also described by Feintuch and Treybal. The method can be applied when overall transfer units can be used and the absorption of each component is described as being either gas-phase or liquid-phase controlled.

While the method of Feintuch and Treybal (1978, 1973) is essentially rigorous and can describe both large heat effects and multicomponent absorption, no thorough experimental comparisons have been made. Kelly et al (May, 1981) have presented some preliminary findings that indicate the algorithm can be used to describe the physical absorption of acid gases from coal gasification product gases. Additional work is necessary to determine the general applicability of the calculational technique.

PACKED ABSORPTION COLUMN MODEL

MODEL DEVELOPMENT

Several assumptions typically have been made in developing a physical model for non-isothermal absorption in a packed tower. Axial dispersion is usually neglected so that fluids are assumed to move through the column in plug flow. This is probably valid for small diameter columns with low to moderate liquid flow rates (Brittan, 1966). Another assumption often made is that the column approaches adiabatic operation; most industrial columns have been shown to operate nearly adiabatically (Bourne, Stockar and Coggan, 1974). Departures from adiabatic operation, due either to heat losses or planned heat removal, can be factored into the model on a case-by-case basis.

Figure 2 shows a schematic of a packed tower for which the following differential material and energy balances may be written.

$$\partial L_j / \partial z - \partial G_j / \partial z = \partial b_j / \partial t \tag{1}$$

$$\partial (LH_L) / \partial z - \partial (GH_G) / \partial z - H_{ex} = \partial (bH_b) / \partial t \tag{2}$$

The resulting system of coupled, partial differential equations is very difficult to solve and several approaches are possible.

As noted in the previous section, Feintuch and Treybal (1978, 1973) suggested a rigorous adiabatic, steady state model of the packed absorber. At steady-state, Equations (1) and (2) become ordinary differential equations

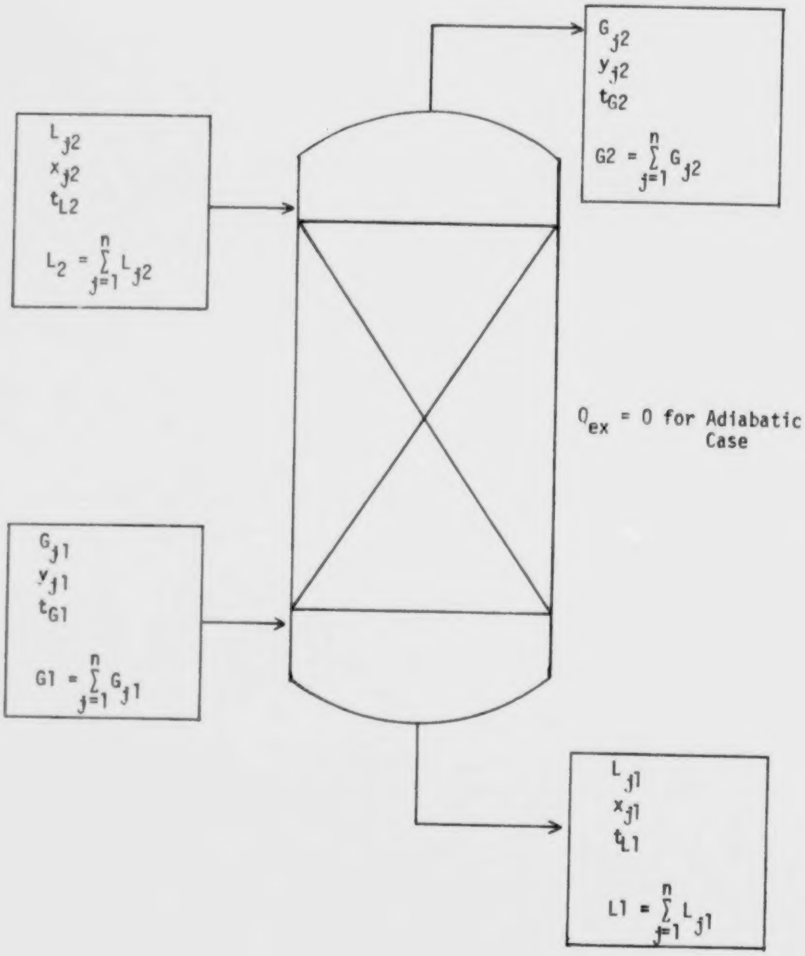
$$dL_j/dz - dG_j/dz = 0 \tag{3}$$

$$d(LH_L)/dz - d(GH_G)/dz = 0 \tag{4}$$

The steady state assumption leads to a two point boundary value problem where neither boundary condition is completely specified. The solution, as suggested by Feintuch and Treybal, involves an iterative calculation in which

Figure 2

MULTICOMPONENT GAS ABSORPTION / STRIPPING IN AN ADIABATIC PACKED COLUMN



L_{j2}
x_{j2}
t_{L2}
$L_2 = \sum_{j=1}^{n} L_{j2}$
G_{j2}
y_{j2}
t_{G2}
$G2 = \sum_{j=1}^{n} G_{j2}$
Q_{ex} = 0 for Adiabatic Case
G_{j1}
y_{j1}
t_{G1}
$G1 = \sum_{j=1}^{n} G_{j1}$
L_{j1}
x_{j1}
t_{L1}
$L1 = \sum_{j=1}^{n} L_{j1}$

the conditions at one end of the column are assumed and an incremental calculation is initiated at the other end. When the calculation reaches the end of the column where the initial assumptions were made, calculated and assumed values are compared. If agreement between these values is unsatisfactory, an iterative procedure must be followed to adjust assumed values. Algorithms have been suggested for both single solute (Treybal, 1969) and multicomponent systems (Feintuch, 1978) although experimental verification of only the former case has been reported (Raal and Khurana, 1973, 1970).

MODEL DESCRIPTION

Aside from the assumptions that the column is operating adiabatically and that axial dispersion is negligible, the calculational procedure first suggested by Treybal (1969) is essentially rigorous. While more simplifying assumptions could be made, such as ignoring the resistance of one phase to mass and heat transfer, this could lead to serious error in the calculation. Certain situations may allow a less rigorous approach but it is often difficult to determine this without performing a more detailed calculation first.

The physical model proposed by Treybal (1969) utilizes the concept of the "two-resistance" theory of gas absorption (Whitman, 1923, 1924). A representation of this model appears in Figure 3. In each differential section of the column, a solute molecule must transfer from the bulk gas phase to the gas-liquid interface, where equilibrium is assumed, and then transfer from the interface into the bulk liquid phase. Solvent evaporation may also occur, as well as the transfer of solutes from the liquid to the gas. Heat transfer is important because of potential differences in gas and liquid temperatures, and because of heat released or absorbed at the interface due to absorption or vaporization. Interfacial areas for mass and heat transfer are assumed to be the same.

While a detailed derivation of the important mathematical expressions used in the calculation is presented by Treybal (1969), a brief summary will be presented here. Nomenclature is given at the end of this report.

The molar flux of component j can be expressed as:

$$\begin{aligned} N_j a\,dz &= R_j F_{G_j} \{\ln[(R_j - y_{ji})/(R_j - y_j)]\} a\,dz = -dG_j \\ &= R_j F_{L_j} \{\ln[(R_j - x_j)/(R_j - x_{ji})]\} a\,dz = -dL_j \end{aligned} \qquad (5)$$

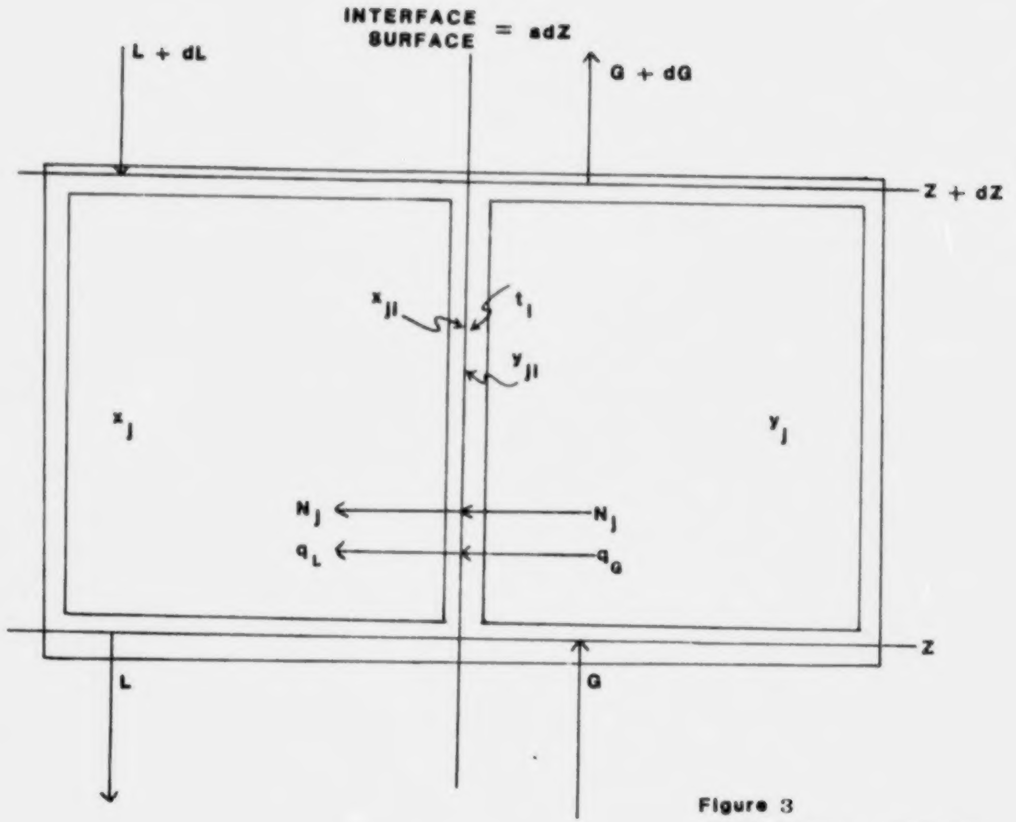

INTERFACE SURFACE = adZ
L + dL
G + dG
Z + dZ
x_ji
t_i
y_ji
x_j
y_j
N_j
N_j
q_L
q_G
Z
L
G

Figure 3
DIFFERENTIAL SECTION

where $R_j = N_j / \Sigma N_j$

and

$$\Sigma R_j = 1.0 \tag{6}$$

From Equations (5) and (6), the gradient of component j in the gas or liquid phase can be computed

$$dG_j/dz = R_j F_{G_j} a\{\ln[(R_j - y_{ji})/(R_j - y_j)]\} \tag{7}$$

$$dL_j/dz = R_j F_{L_j} a\{\ln[(R_j - x_j)/(R_j - x_{ji})]\} \tag{8}$$

The composition of the gas or liquid phase at the interface can be calculated from the phase equilibrium relationships and by combining Equations (5) and (6) to solve for y_{ji} or x_{ji}.

$$y_{ji} = f\ (T,P,x_{ji}) \tag{9}$$

$$x_{ji} = f\ (T,P,y_{ji}) \tag{9a}$$

$$y_{ji} = R_j - (R_j - y_j)[(R_j - x_j)/(R_j - x_{ji})]^{F_{L_j}/F_{G_j}} \tag{10}$$

$$x_{ji} = R_j - (R_j - x_j)[(R_j - y_{ji})/(R_j - y_j)]^{F_{L_j}/F_{G_j}} \tag{10a}$$

Energy balances are necessary to determine temperature gradients and the interfacial temperature, which is not necessarily the same as that of the bulk liquid. The gas phase enthalpy and its differential can be expressed as

$$H_G = \Sigma G_j\ (C_{Gj}\ [t_G - t_o] + \lambda_j) \tag{11}$$

$$dH_G = \Sigma G_j\ dt_G + (C_{Gj}\ [t_G - t_o] + \lambda_j)\ dG_j \tag{12}$$

The liquid phase enthalpy and its differential are as follows

$$H_L = \Sigma L_j\ (C_{Lj}\ [t_L - t_o] + \Delta H_{Sj}) \tag{13}$$

$$= \Sigma L_j \, (C_{Gj} \, [t_L - t_o] + \lambda_j + \Delta H_{Sj}) \tag{13a}$$

$$dH_L = \Sigma L_j \, (dt_L + d \, \Delta H_{Sj}) + (C_{Gj} \, [t_L - t_o] + \lambda_j + \Delta H_{Sj}) \, dL_j \tag{14}$$

Heat transfer rates between the gas and liquid phases per unit interfacial area are defined in terms of gas and liquid-phase heat transfer coefficients, h_G and h_L.

$$q_G \, a \, dz = h_G a \, [t_G - t_o] \, dz \tag{15}$$

$$q_L \, a \, dz = h_L a \, [t_L - t_o] \, dz \tag{16}$$

The bulk gas temperature gradient can be determined by combining Equations (5), (11), and (12):

$$dt_G/dz = h_G a [t_G - t_i] \; / \; \Sigma G_j \, C_{Gj} \tag{17}$$

The interfacial temperature can be determined from the above information as described by Treybal (1969):

$$t_i = (1.0 \, / \, h_L a) \;\; \Sigma [H_{ji} - (C_{Gj} [t_G - t_o] + \lambda_j)] \, dG_j/dz + t_L - (1.0 \, / \, h_L a) \, dt_G/dz \;\; \Sigma G_j C_{Gj} \tag{18}$$

where H_{ji} = partial molar enthalpy of component j in solution at x_{ji} and t_i

These equations are then used in an appropriate algorithm to determine the height of packing required for the specified recovery of a key solute, in case of a design statement, or the recovery of a specified solute for a given height of packing, in the case of an analysis statement.

DETERMINATION OF PACKED HEIGHT

The algorithm proposed by Feintuch and Treybal (1978, 1973) was formulated as a design statement. In this algorithm, Equation 3 is modified to give

$$dG_j = dL_j \tag{19}$$

From Equations (5) and (6)

$$dz = - \{1/R_j F_{G_j} a \ln[(R_j - y_{ji})/(R_j - y_j)]\} dG_j \tag{20}$$

$$dz = - \{1/R_j F_{L_j} a \ln[(R_j - x_j)/R_j - x_{ji})]\} dL_j \tag{21}$$

In essence, either equation can be used to determine the required height of packing for the recovery of any solute, j. However, either solution must be obtained by including the mass transfer rate expressions for all components, the phase equilibrium relationships, the heat transfer rate expressions for each phase, and the material and energy balances for a differential section. Because of the implicit, non-linear nature of the resulting system of equations, an analytical solution is not available and the integral must be evaluated numerically.

The algorithm suggested by Feintuch and Treybal (1978) attempts to evaluate the integral by summing the heights required to affect a specified change in component G_j for a previously selected number of packing segments. In each segment, it is necessary to determine the interfacial composition and temperature as well as the molar flux of each component with an iterative procedure. Temperature and concentration gradients are then combined with material and energy balances to calculate the composition, temperature and flow rate of both phases in the adjacent packing segment.

A more detailed description of the algorithm is provided in the next section.

DESCRIPTION OF COMPUTER PROGRAMS

Two separate but similar computer programs were used in this study. One, SIMPAK, was limited to the simple case of a single transferring solute in an inert, insoluble carrier gas and a volatile solvent. The other, MCOMP, relaxed these restrictions. Both will be discussed in this section.

SIMPAK

This program is a modification of computer codes presented by Feintuch (1973) and Khurana (1970). Because mass transfer is limited to two components, the model is greatly simplified. First, since the sum of the normalized molar fluxes must be unity

$$R_{key} + R_{solv} = 1.0 \tag{22}$$

Second, since the inert carrier gas will not be present in the liquid phase

$$x_{key} + x_{solv} = 1.0 \tag{23}$$

SIMPAK receives as input the condition and composition of the entering gas and liquid streams and the specified removal efficiency of the key component (see Table 1). Also entered into the program are the number of packing segments or step size, the chosen transport property and coefficient options, and the adjustable interfacial area parameter. The key component can be chosen from the list of 13 compounds shown in Table 2. Methanol, the liquid used in this study, must be designated as the solvent. The inert carrier gas can also be any compound in this list except for methanol. In general, the key component selected should be one that is appreciably soluble in methanol while the inert component should be a compound that is sparingly soluble in methanol.

Figures 4 and 5 illustrate the iterative procedure used in SIMPAK. Since the boundary conditions at both ends of the packing are not completely specified, one set of conditions must be assumed in order to initiate the calculation. In this case, the outlet conditions at the top of the tower are assumed which allows the boundary conditions at the bottom of the tower to be determined by material and energy balances. This approach is taken because conditions of the exit gas, especially its enthalpy, are more easily

TABLE 1

Packing Height For Run AM-32 Simpak Test 07/12/81 11:00

Gas Mass Vel. Mol/Hr/Sq.Ft	Gas Temp. In Deg. F	Liq. Mass Vel. Mol/Hr/Sq.Ft	Liq. Temp. In Deg. F
17.31	54.42	61.81	-31.79

Pressure Atmos.	The Key Comp.	Fract Key Comp. Absorbed
28.210	1	0.981000

Comp. No.	Comp.	Inlet Gas Mol.Fract	Inlet Liquid Mol.Fract
1	CO2	0.3233	0.0000
2	H2S	0.0000	0.0000
3	COS	0.0000	0.0000
4	MEOH	0.0000	1.0000
5	H2	0.0000	0.0000
6	CO	0.0000	0.0000
7	N2	0.6767	0.0000
8	CH4	0.0000	0.0000
9	C2H4	0.0000	0.0000
10	C2H6	0.0000	0.0000
11	C3H6	0.0000	0.0000
12	C3H8	0.0000	0.0000
13	BUT	0.0000	0.0000

Binary Gas Diffusivity Option: Wilke-Lee Modification to Chapman-Enskog

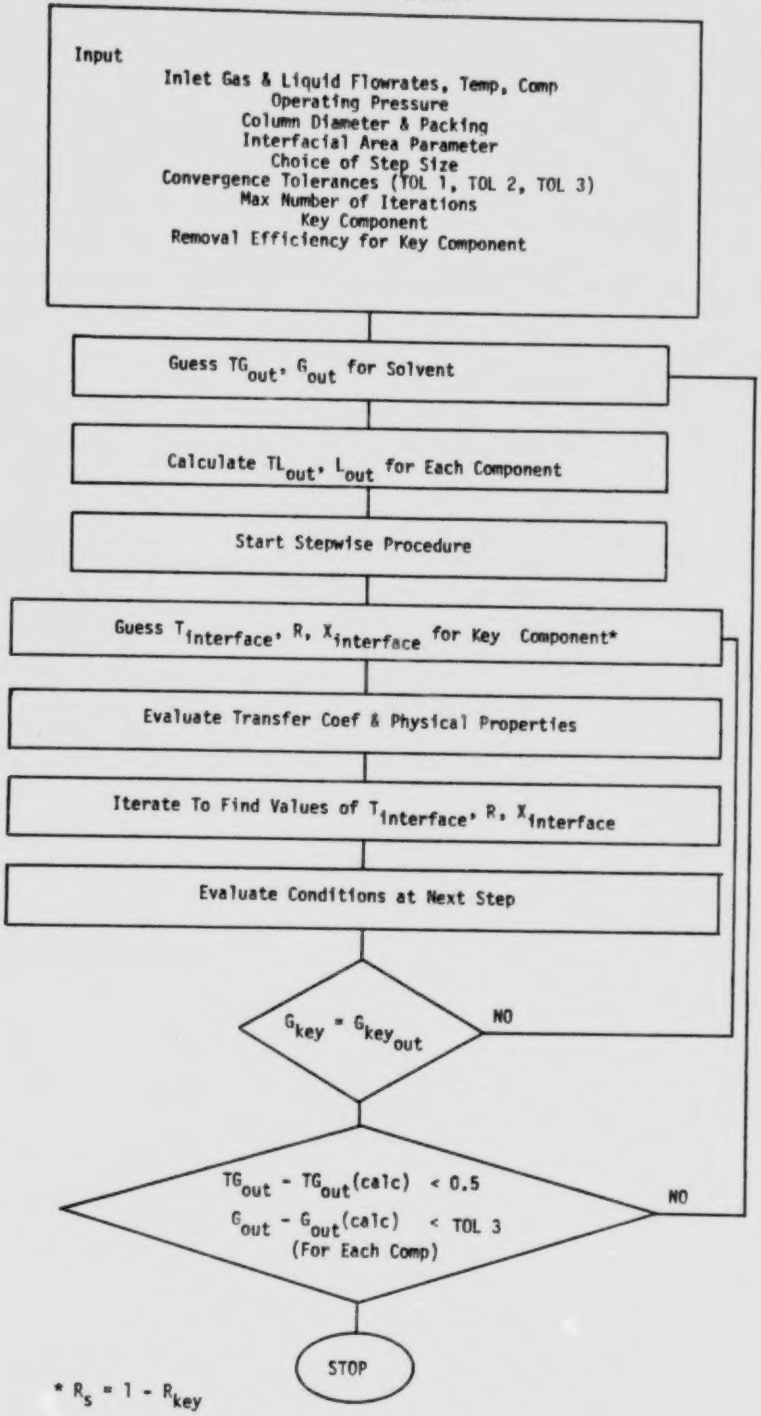
Binary Liquid Diffusivity Option: Wilke-Chang Modification to Stokes-Einstein EQN

Transfer Coefficient Option: Onda Et Al.

Packing Height	7.10 Feet
Packing Type	1/4 Inch Ceramic Intalox Saddles
Packing Increm	10.0
Int. Area Corr.	1.00
Maximum Iterations	5

Figure 4

SIMPAK FLOWCHART


Input
Inlet Gas & Liquid Flowrates, Temp, Comp
Operating Pressure
Column Diameter & Packing
Interfacial Area Parameter
Choice of Step Size
Convergence Tolerances (TOL 1, TOL 2, TOL 3)
Max Number of Iterations
Key Component
Removal Efficiency for Key Component
Guess TG_{out}, G_{out} for Solvent
Calculate TL_{out}, L_{out} for Each Component
Start Stepwise Procedure
Guess $T_{interface}$, R, $X_{interface}$ for Key Component*
Evaluate Transfer Coef & Physical Properties
Iterate To Find Values of $T_{interface}$, R, $X_{interface}$
Evaluate Conditions at Next Step
$G_{key} = G_{key_{out}}$
NO
$TG_{out} - TG_{out}(calc) < 0.5$
$G_{out} - G_{out}(calc) < TOL\ 3$
(For Each Comp)
NO
STOP

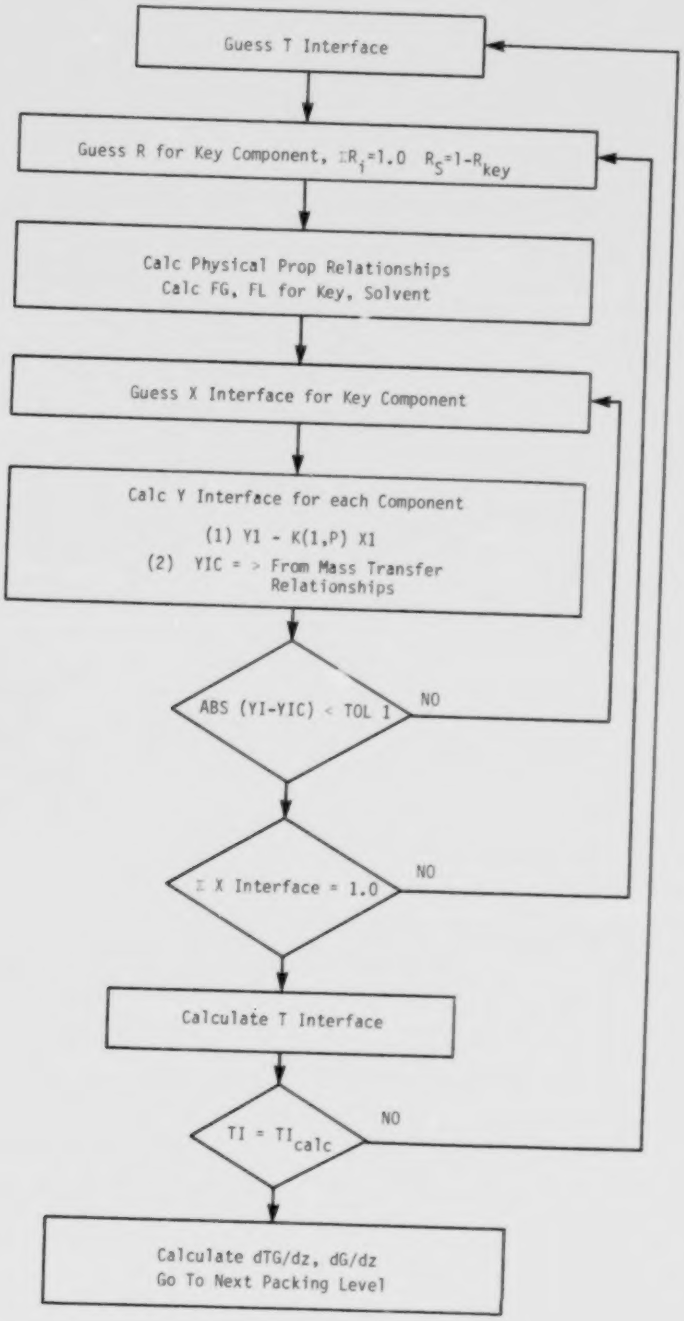
* $R_s = 1 - R_{key}$

$X_s = 1 - X_{key}$

Figure 5

SIMPAK

ITERATIVE PROCEDURE IN EACH DIFFERENTIAL SECTION


Guess T Interface
Guess R for Key Component, $\Sigma R_i=1.0$ $R_S=1-R_{key}$
Calc Physical Prop Relationships
Calc FG, FL for Key, Solvent
Guess X Interface for Key Component
Calc Y Interface for each Component
(1) YI - K(I,P) XI
(2) YIC = > From Mass Transfer Relationships
ABS (YI-YIC) < TOL 1
NO
Σ X Interface = 1.0
NO
Calculate T Interface
TI = TI_{calc}
NO
Calculate dTG/dz, dG/dz
Go To Next Packing Level

estimated than are those of the exit liquid.

As a first assumption, the outlet gas temperature is set equal to the temperature of the incoming solvent and the outlet gas is assumed to be saturated with the solvent at this temperature. Because the removal efficiency of the key component has been specified, the boundary conditions at the top of the tower are then defined. With this information the temperature, composition and flow rate of the liquid leaving the column can be determined by material and energy balances.

In the numerical algorithm, the column has been divided into a finite number of segments in which equal amounts of the key component are to be removed; the calculational procedure must determine the height required to affect the specified removal in each segment. Because driving forces for heat and mass transfer will vary along the column, the height required in each segment, in general, will be different.

TABLE 2

COMPONENTS INCLUDED IN MODEL
CARBON DIOXIDE
HYDROGEN SULFIDE
CARBONYL SULFIDE
METHANOL
HYDROGEN
CARBON DIOXIDE
NITROGEN
METHANE
ETHYLENE
ETHANE
PROPYLENE
PROPANE
BUTANE

In each packing segment, an iterative procedure is required to determine the concentration and temperature gradients needed to compute the conditions at the next packing level. This procedure is illustrated in Figure 5 and described in detail by Treybal (1969). Once the gradients are determined, the following equations are used to determine the packing height required in that segment as well as the conditions at the next packing level:

$$Z_{next} = Z + \Delta Z \tag{24}$$

$$= Z + \Delta G_{key}/[dG_{key}/dZ]$$

$$G_{key,next} = G_{key} + \Delta G_{key} \tag{25}$$

$$G_{solv,next} = G_{solv} + \Delta G_{solv} \tag{26}$$

$$= \Delta Z[dG_{solv}/dZ]$$

$$t_{G,next} = t_G + \Delta Z[dt_G/dZ] \tag{27}$$

$$L_{next} = L + \Delta G_{key} + \Delta G_{solv} \tag{28}$$

$$x_{key,next} = (\Delta G_{key} + L_{key})/L_{next} \tag{29}$$

$$H_{L,next} = \{(LH_L)_{top} + (H_{G,next} - H_{G,top})G\}/L_{next} \tag{30}$$

The concentration of the solute at the next packing level is compared to that specified in the program input to see if the top of the tower has been reached. If the top of the tower has not been reached, the stepwise calculation continues; if it has been reached, the assumed values of the outlet gas temperature and solvent concentration are compared to the calculated values. The iteration continues if these values do not agree within a specified tolerance using the calculated values for the next assumptions of outlet gas temperature and composition.

The following subroutines comprise the computer program SIMPAK. All subroutines are written in FORTRAN and are given by Kelly (1981). A brief description of each is presented here:

SIMPAK	Drives the computer program and coordinates all subroutines.
INPUT	Reads in all input data, as well as physical and transport property correlation options.
ENTHAL	Calculates the enthalpies of all inlet and outlet flow streams.
VLE	Contains all vapor-liquid phase equilibrium information.

HCAP	Calculates individual and mixture heat capacities for gas and liquid phases.
HSOLN	Determines heats of solution from Henry's Law constants.
MIXDIF	Calculates mixture diffusivities and heat and mass transfer coefficients.
TEMPI	Coordinates iterative calculation in each differential section.
NSTEP	Calculates information on conditions at next packing level.
VISLG	Calculates individual and mixture viscosity for gas and liquid phases.
RHO	Calculates individual and mixture densities for gas and liquid phases.
TCON	Calculates individual and mixture conductivities for gas and liquid phases.
PACK1	Determines interfacial area for heat and mass transfer.
DIFFG	Calculates binary diffusivities for gas phase components.
DIFFL	Calculates binary diffusivities for liquid phase components.
SOLV	Performs linear interpolation.

Details on the content of these subroutines and the numerical characteristics of the algorithm are discussed in a later section.

MCOMP

For the case of multicomponent absorption, a computer program called MCOMP has been developed. The program is a modification of the computer code used by Feintuch (1973). Although any number of components can be handled, the thirteen components designated as the most important in coal gasification applications have been made a part of the model. A list of these compounds is given in Table 2.

Because any of the thirteen components can transfer in either direction, the problem becomes much more complicated than that for single solute absorption. Although the algorithm used in this case is similar to SIMPAK, inclusion of multicomponent mass transfer requires certain modifications. Flow charts of the calculations are shown in Figures 6 and 7.

After input of all necessary information (see Table 3), the calculation is initiated by assuming the outlet gas temperature and outlet gas flow rates for all components, except for the key component, which has been specified. The outlet gas temperature is assumed initially to be equal to the inlet liquid temperature. An initial estimate of the outlet gas flow rates is obtained by taking into account the Henry's law constant of a particular compound relative to the key component at the assumed temperature of the outlet gas. For compounds less soluble than the key component, the assumed value of the outlet flow rate is determined with the following expression:

$$G_{j,\mathrm{assumed}} = 0.99[1.0-(\mathrm{SPEC})(\mathrm{RATIO}_j)](\mathrm{VMG})(\mathrm{XMOLG}_j) \tag{31}$$

where G_j = assumed outlet flow rate of component j

SPEC = removal efficiency of the key component

RATIO_j = ratio of Henry's law constant of the key component to component j

VMG = inlet molar gas flow rate

XMOLG_j = inlet gas mole fraction of component j

For compounds more soluble than the key component, the following expression is used:

$$G_{j,\mathrm{assumed}} = (1.0-\mathrm{SPEC})(\mathrm{VMG})(\mathrm{XMOLG}_j)/\mathrm{RATIO}_j \tag{32}$$

After the flow rates of all exiting gas stream components are assumed, material and energy balances are solved to determine the conditions of the exiting liquid stream.

The calculation then begins at the bottom of the column. The total amount of the key component to be removed in the column is divided by the number of packing segments to be used. The height required to achieve the removal of the key component in each segment is determined by calculating the temperature and concentration gradients as was done in SIMPAK. The technique used to determine the gradients in each differential section is, however, different and will be described here.

Figure 7 shows the iterative procedure used in each differential section to determine the composition and temperature gradients in MCOMP. After making an initial estimate of the interfacial temperature (usually two

Figure 6

MCOMP FLOWCHART

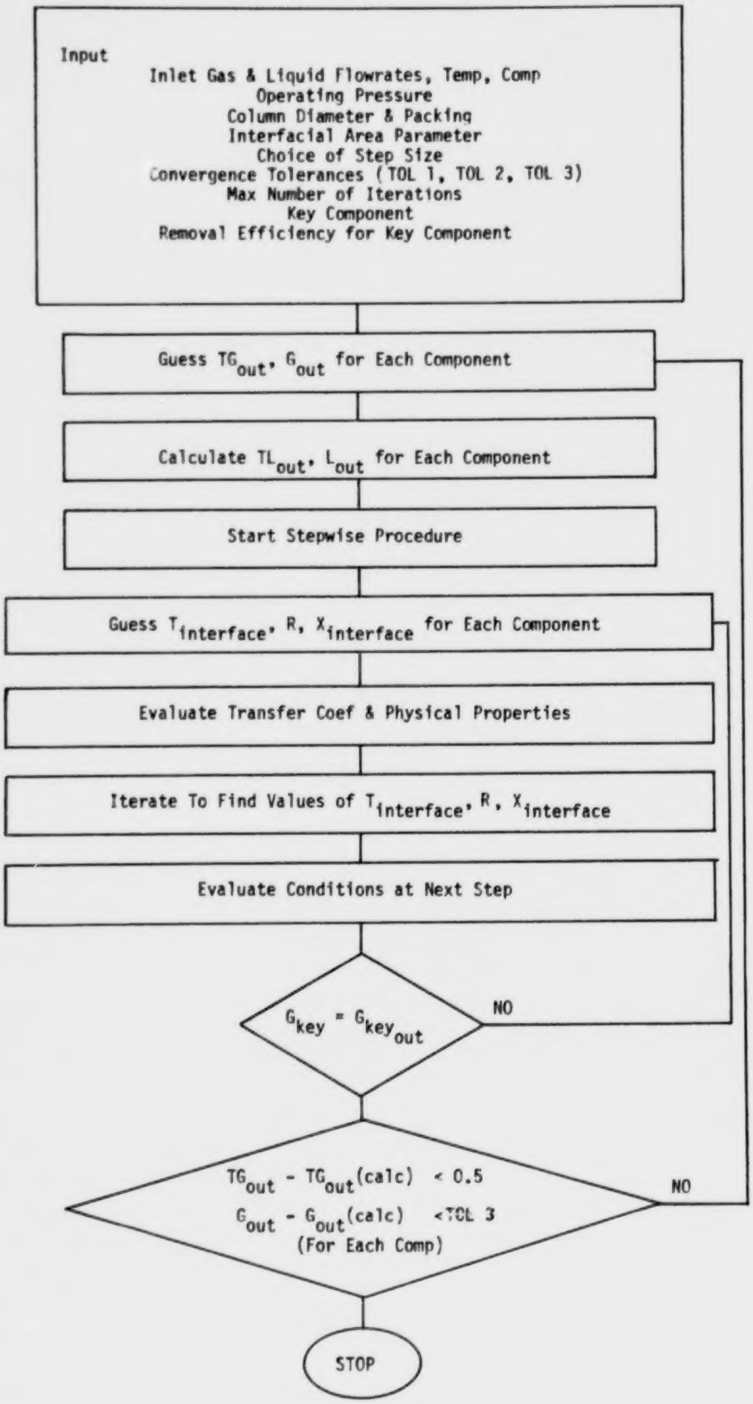

Input
Inlet Gas & Liquid Flowrates, Temp, Comp
Operating Pressure
Column Diameter & Packing
Interfacial Area Parameter
Choice of Step Size
Convergence Tolerances (TOL 1, TOL 2, TOL 3)
Max Number of Iterations
Key Component
Removal Efficiency for Key Component
Guess TG_out, G_out for Each Component
Calculate TL_out, L_out for Each Component
Start Stepwise Procedure
Guess T_interface, R, X_interface for Each Component
Evaluate Transfer Coef & Physical Properties
Iterate To Find Values of T_interface, R, X_interface
Evaluate Conditions at Next Step
G_key = G_key_out
NO
TG_out - TG_out(calc) < 0.5
G_out - G_out(calc) <TOL 3
(For Each Comp)
NO
STOP

Figure 7

MCOMP

ITERATIVE PROCEDURE IN EACH DIFFERENTIAL SECTION

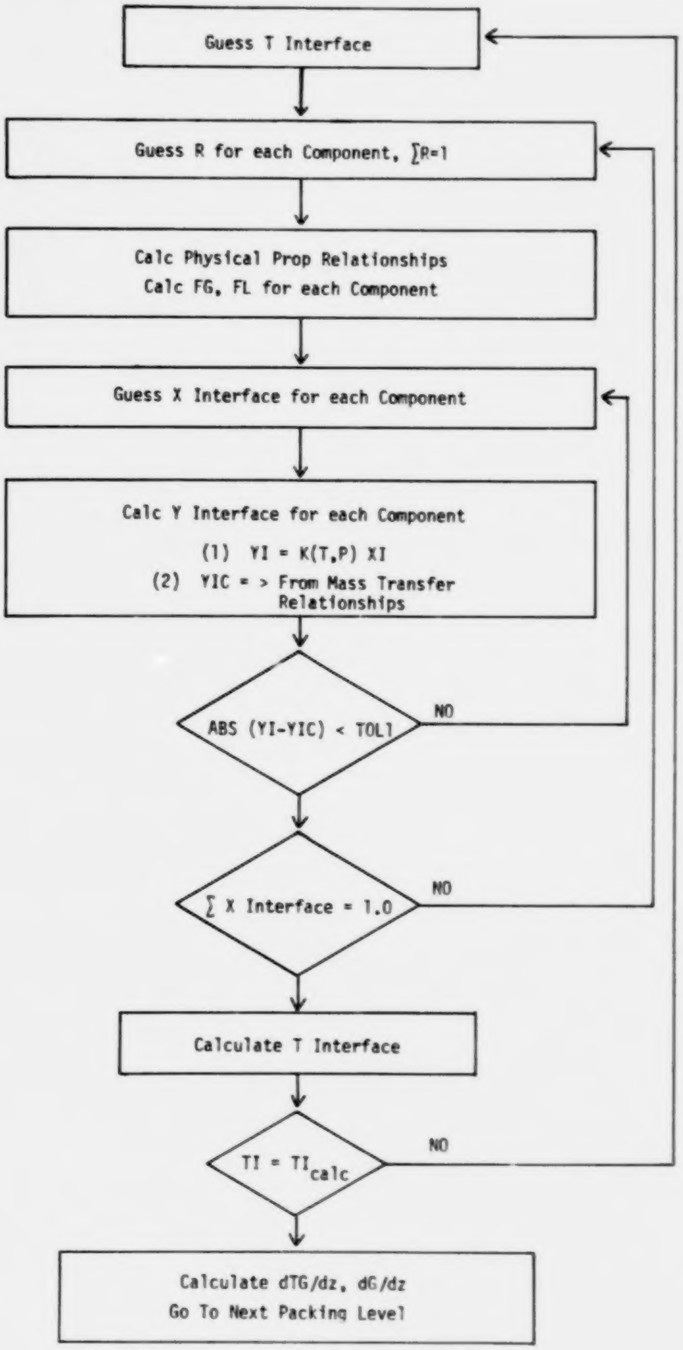

Guess T Interface
Guess R for each Component, ∑R=1
Calc Physical Prop Relationships
Calc FG, FL for each Component
Guess X Interface for each Component
Calc Y Interface for each Component
(1) YI = K(T,P) XI
(2) YIC = > From Mass Transfer Relationships
ABS (YI-YIC) < TOL1
NO
∑ X Interface = 1.0
NO
Calculate T Interface
TI = TI calc
NO
Calculate dTG/dz, dG/dz
Go To Next Packing Level

TABLE 3

Run AMI-53 MCOMP Simulation 08/11/81 11:30

Gas Mass Vel. Mol/Hr/Sq.Ft	Gas Temp. In Deg. F	Liq. Mass Vel. Mol/Hr/Sq.Ft	Liq. Temp. In Deg. F
17.88	42.53	127.46	-4.28

Pressure Atmos.	The Key Comp.	Fract Key Comp. Absorbed
17.890	1	0.99900

Comp. No.	Comp.	Inlet Gas Mol.Fract	Inlet Liquid Mol.Fract
1	CO2	0.2213	0.0000
2	H2S	0.0017	0.0000
3	COS	0.0001	0.0000
4	MEOH	0.0000	1.0000
5	H2	0.3292	0.0000
6	CO	0.1724	0.0000
7	N2	0.2112	0.0000
8	CH4	0.0529	0.0000
9	C2H4	0.0045	0.0000
10	C2H6	0.0065	0.0000
11	C3H6	0.0000	0.0000
12	C3H8	0.0000	0.0000
13	C4H10	0.0000	0.0000

Binary Gas Diffusivity Option: Wilke-Lee Modification to Chapman-Enskog

Binary Liquid Diffusivity Option: Wilke-Chang Modification to Stokes-Einstein EQN

Mixture Liquid Diffusivity Option: Modification of Holmes et al. to Wilke-Chang Equation

Transfer Coefficient Option: ONDA et al.

Packing Height	7.10 Feet
Packing Type	1/4 Inch ceramic intalox saddles
Packing Increm	10.0
Int. Area Corr.	1.00
Maximum Iterations	0
Convergence Toleranc	0.0000100 0.0001000 0.0150000 0.0050000
Convergence Accelera	0.50000 0.50000

degrees above the bulk liquid temperature), values of the normalized fluxes of each transferring component are assumed subject to the following restriction:

$$\Sigma R_j = 1.0 \tag{33}$$

Initially, all fluxes are assigned values of 1.0 except for the solvent which is equal to

$$R_{solv} = 1.0 - \text{number of components} \tag{34}$$

Next, the liquid composition at the interface in the first packing segment is assumed to be

$$x_{j,interface} = 1.05\ x_j \tag{35}$$

with the solvent fraction set equal to the difference between the sum of the other component fractions and one.

For the assumed values of T(interface) and the normalized molar fluxes, R_j, the intersection of Equations (9) and (10) is determined iteratively for each component using linear interpolation to refine the assumed values of x_j (interface). When the equations have been satisfied to within a given tolerance, the values of x_j (interface) are summed. If this sum is within a given tolerance of 1.0, the interfacial temperature is calculated using Equation (18) and compared to the assumed value. If the agreement is satisfactory, the gas phase gradients for all components and the gas temperature gradient are calculated using Equations (7) and (17). These are used to determine the conditions at the next packing level with the following expressions:

$$\begin{aligned} Z_{next} &= Z + \Delta Z \\ &= Z + \Delta G_{key}/[dG_{key}/dZ] \end{aligned} \tag{24}$$

$$G_{key,next} = G_{key} + \Delta G_{key} \tag{25}$$

$$\begin{aligned} G_{j,next} &= G_j + \Delta G_j \\ &= \Delta Z[dG_j/dZ] + G_j \end{aligned} \tag{36}$$

$$t_{G,next} = t_G + \Delta Z[dt_G/dZ] \tag{27}$$

$$L_{next} = L + \Delta G_j \tag{37}$$

$$x_{j,next} = [\Delta G_j + L_j]/L_{next} \tag{38}$$

$$H_{L,next} = \{(LH_L)_{top} + (H_{G,next} - H_{G,top})G\}/L_{next} \tag{30}$$

At the next packing level a check is made to see if the top of the column has been reached. If it has not, the calculation continues. If it has been reached, as determined by comparing the calculated and specified outlet gas flow rate of the key component, all of the other outlet gas flow rates are compared to the assumed values. When these values agree to within a prescribed tolerance, the assumed and calculated outlet gas temperatures are compared to see if the computation has converged. If convergence is not achieved, new values of the outlet gas flow rates are made by using weighted successive substitution as suggested by Feintuch (1973):

$$G_{j,new} = 0.5(G_{j,old} + G_{j,calc}) \tag{39}$$

The new outlet gas temperature is equal to the most recently calculated value.

If the sum of the interfacial liquid mole fractions is not close enough to one, new values of the normalized molar fluxes must be chosen. Using the method suggested by Feintuch (1973), a new value of the normalized flux of component j can be determined by weighted successive substitution:

$$N_j = R_j F_{G_j} \ln[(R_j - y_{j,interface})/(R_j - y_{j,bulk})] \tag{40}$$

$$R_{j,new} = 0.5\ \{[N_j/\ \Sigma N_j] + R_{j,old}\} \tag{41}$$

After the new values of R_j have been determined, physical and transport properties and coefficients are calculated and the iteration continued.

The following subroutines are contained in the program MCOMP. All subroutines are written in FORTRAN and are given by Kelly (1981). Several of these are essentially the same as those used in SIMPAK. A brief description of each is presented here:

MCOMP	Drives the computer program and coordinates all subroutines.
INPUTM	Reads in all input data, as well as transport property and coefficient correlation options.
ENTHM	Calculates the enthalpy of all inlet and outlet flow streams.
VLEMLT	Contains all vapor-liquid phase equilibrium information.
HCAPM	Calculates individual and mixture heat capacities for gas and liquid phases.
HSOLNM	Determines heats of solution from Henry's Law Constants.
MLTDIF	Calculates mixture diffusivities and heat and mass transfer coefficients.
MTEMPI	Coordinates iterative calculation in each differential section.
NSTEPM	Calculates information on conditions at next packing level.
VISLGM	Calculates individual and mixture viscosities for gas and liquid phases.
RHOMLT	Calculates individual and mixture densities for gas and liquid phases.
PACK1M	Determines interfacial area for heat and mass transfer.
DIFFGM	Calculates binary diffusivities for gas phase components.
DIFFLM	Calculates binary diffusivities for liquid phase components.
TCONM	Calculates individual and mixture thermal conductivities for gas and liquid phases.
SOLV	Performs linear interpolation and extrapolation.

Details on the content of these subroutines are provided later. The numerical characteristics of the algorithm are also discussed later.

PHYSICAL, TRANSPORT, AND THERMODYNAMIC PROPERTIES

The success of any modeling effort hinges on either the availability of accurate physical, transport, and thermodynamic property data or the ability to predict this information. Suprisingly, relatively little effort has been invested in expanding existing physical property data bases and improving predictive methods. This is especially true for mixture properties and at other than ambient conditions.

Built into both SIMPAK and MCOMP are subroutines that compute mixture physical property information as needed in the model calculation. A brief description of how these properties were determined is provided below. Further details can be found in the computer codes in the Appendices.

VISCOSITY

Both gas and liquid viscosities were computed in the subroutines VISLG (SIMPAK) and VISLGM (MCOMP). Single component gas viscosities are correlated as functions of temperature from data available in the literature. Viscosity of gas mixtures was calculated using Wilke parameters as suggested by Reid et al (1977) and the pressure correction of Dean and Steil (1963). It is recognized that the presence of large amounts of hydrogen may lead to some inaccuracy but no available method handles this situation.

Liquid viscosities were computed for pure components from the data found in Reid et al (1977). Viscosity of liquid mixtures was calculated using the relationship found in the Perry and Chilton Handbook (1973). Deviations in the mixture viscosity resulting from heat of mixing effects were ignored.

THERMAL CONDUCTIVITY

Gas and liquid thermal conductivities were computed in subroutines TCON (SIMPAK) and TCONM (MCOMP). Thermal conductivities for pure gases were correlated as functions of temperature from data available in the literature. Thermal conductivity for gas mixtures was determined using the mixture rule found in the Perry and Chilton Handbook (1973). Although there are problems using this mixture rule with hydrogen and polar compounds, no other method was available. The effect of pressure on gas thermal conductivities was accounted for with the pressure correction method of Stiel and Thodus (1964).

Thermal conductivities of liquids were expressed as functions of temperature by equations resulting from least squares fits of available data. The assumption was made that only methanol and carbon dioxide contribute to the liquid thermal conductivity, since the other components are usually present in very small amounts. Methanol thermal conductivity shows a linear relationship with temperature from 32°F to 122°F. It was assumed that this relationship can be extrapolated to lower temperatures where no data are available. Because the temperature range of interest brackets the critical temperature of CO_2, a simple linear relationship with temperature is not sufficient. The thermal conductivity over this range shows a maximum and, therefore, a cubic polynomial was used to describe the data.

The mixture thermal conductivity was calculated using the Power Law Method of Vredeveld as recommended in Reid et al (1977). As suggested, an exponent of $R=-2.0$ was used. An exponent of $R=1.0$ would provide a weight fraction average.

HEAT CAPACITY

Heat capacity information is determined in subroutines HCAP (SIMPAK) and HCAPM (MCOMP). Gas phase heat capacities were assumed to be independent of pressure. Their variation with temperature was determined from information available in Reid et al (1977). The mole fraction-weighted average of these values was used as the mixture value.

Because only methanol and carbon dioxide are present in any appreciable amount in the liquid phase, only their heat capacities were taken into account in calculating the liquid mixture heat capacity. The mixture heat capacity was taken as the mole fraction-weighted average of the two components at the particular temperature of interest. The data for liquid CO_2 heat capacity are reported at pressures of 63 atmospheres; extension of this information to the pressure range of interest was done by ignoring the effects of pressure on liquid properties.

The liquid mixture weighting procedure is subject to error especially since there are large heat effects. However, no other method is available to account for the influence of these effects on the mixture heat capacity.

DENSITY

Gas and liquid densities are computed in subroutines RHO (SIMPAK) and RHOMLT (MCOMP). The gas phase density was computed by assuming the gas phase behaves ideally. This assumption is consistent with the use of Henry's law to express the phase equilibrium relationships. Some inaccuracy is expected and can be reduced in the future by incorporating the equations of state used by Rousseau et al (May, 1980, January, 1980) into the model.

Liquid density was calculated by assuming that only methanol and CO_2 contribute to the mixture density. The liquid density of these two components were determined from the relationships found in Prausnitz et al (1969) and the mole fraction-weighted average of the two was used to determine the mixture value. It was assumed that there is no volume change upon mixing.

VAPOR-LIQUID PHASE EQUILIBRIUM RELATIONSHIPS

Phase equilibrium relationships are required for quantitative design or analysis of any mass transfer operation. These relationships are often unavailable because of difficulties associated with measurement and/or prediction of this information.

In this study, several simplifying approximations have been made because of the need to deal with a system having thirteen components. The gas phase has been assumed to be ideal, despite the fact that pressures in excess of 30 atmospheres were used, and Henry's law has been assumed valid for the liquid phase. Henry's law constants were determined from information available in the literature (Lazalde-Crabtree, 1980; Landolt-Bornstein, 1976) and are given as functions of temperature in Table 4.

TABLE 4

HENRY'S LAW CONSTANTS IN METHANOL

$$\mathrm{LN}\ [\mathrm{HENRY\ (BARS)}] = A0 + A1/T(K) + A2/T(K)^{2.0}$$

COMPOUND	A0	A1	A2	TEMPERATURE RANGE(K)
CO2	7.9008	-213.290	-2.107E5	198.15 - 298.15
H2S	11.3789	-2285.560	0.0	193.15 - 303.15
COS	10.5322	-1979.989	0.0	233.15 - 283.15
H2	7.4133	399.818	0.0	213.15 - 313.15
CO	8.6861	-238.794	0.0	193.15 - 313.15
N2	6.7094	801.358	-1.0454E5	213.15 - 298.15
CH4	8.7260	-494.180	0.0	213.15 - 323.15
C2H4	10.1494	-1378.48	0.0	233.15 - 283.15
C2H6	11.3411	-1665.34	0.0	193.15 - 298.15
C3H6	13.3630	-2666.67	0.0	233.15 - 283.15
C3H8	4.6750	1513.57	-4.6805E5	228.15 - 313.15
C4H10	11.3308	-2343.78	0.0	233.15 - 283.15

The simplifying approximations described in the preceeding paragraph will be removed upon the development of more sophisticated descriptions of vapor-liquid behavior. Rousseau et al (1980a, 1980b) have developed a model that predicts phase equilibrium behavior for the systems containing N_2-CO_2-H_2S-Methanol. Work is underway now to expand this model to handle additional components.

Methanol loss is another important factor in the operation of the absorber. Prausnitz et al (1976) have suggested a technique to predict this quantity from phase equilibrium information. In the study, the methanol concentration in the vapor phase was determined from vapor pressure information alone. A more exact approach will be implemented in the future if the experimental results warrant.

In the subroutines VLE (SIMPAK) and VLEMLT (MCOMP), vapor-liquid equilibria are reported as K-values using the following definitions:

$$y_{j,\text{interface}} = K_j \; x_{j,\text{interface}} \qquad (42)$$

where $K_j = \text{Henry}_j/P$

The mole fraction of methanol in the vapor phase was approximated from the vapor pressure of methanol at the system temperature, p^*.

$$y_{\text{methanol}} = p^* \text{ (methanol at T)}/P \qquad (43)$$

This simple approach has proved to be adequate for correlation of experimental results, probably due to the low concentrations of methanol in the vapor phase and gases in the liquid phase at the temperatures and pressures of interest.

Evidence of non-ideality in these systems has been reported. For example, the effect of the presence of H_2 on the solubility of CO_2 has been cited by Ranke (1972) although the extent of this effect has not been documented.

HEAT OF SOLUTION

Absorption of large amounts of carbon dioxide and hydrogen sulfide from coal gas results in the release of significant amounts of heat. Relating this exotherm to the rate of absorption was required in the development of an absorber model. In this regard, gas absorption can be viewed as taking this process path (Prausnitz, 1958): first, the gas condenses and, second, the condensed solute mixes with the solvent. Using this same model, the heat of solution can be viewed as the combination of the heat released in each step: the heat of condensation and the heat of mixing.

There are a number of ways in which the heat of solution can be estimated. Most simply, only the heat of condensation could be considered. This will not suffice for the acid gases CO_2, COS, and H_2S. Another possibility is the use of reference substance plots as suggested by Othmer (1968).

Because Henry's law constants have already been computed as functions of temperature, it was decided to use an approach suggested by Denbigh (1971) which uses these values to estimate heats of solution at infinite dilution. For an ideal vapor phase, the following expression can be derived:

$$d \ln(p_j/x_j) = [(h_j-H_j)/RT^2]\, dT + (V_j/RT)\, dP \qquad (44)$$

where p_j = partial pressure (use fugacity for nonideal gases)
x_j = liquid mole fraction for component j
h_j = enthalpy per mole of gaseous component j
H_j = partial molar enthalpy of component j in solution
R = gas constant
T = absolute temperature
V_j = partial molar volume of component j
P = system pressure

Recognizing that p_j/x_j is the Henry's law constant and that $\{h_j-H_j\}$ represents the heat absorbed in the evaporation of one mole of component j from the ideal solution at constant at temperature and pressure

$$(\partial \ln \text{Henry}_j / \partial T)_P = (h_j - H_j)/RT^2 \qquad (45)$$

or

$$h_j - H_j = RT^2(\partial \ln \text{Henry}_j / \partial T)_P \qquad (46)$$

The left hand term is the heat of solution at infinite dilution.

The expressions presented in Table 4 for Henry's law constants as functions of temperature were used to determine the heat of solution at infinite dilution for each of the components used in the model. The heat of solution of methanol was taken to be its heat of condensation. The influence of the presence of another compound on the heat of solution of a component was ignored.

DIFFUSIVITIES

Diffusivity depends upon temperature and pressure of the system as well as on the nature of the components; it refers to the net transport of material within a single phase in the absence of convective mixing. All procedures for estimation of mass transfer coefficients require a knowledge of diffusivity. Despite the importance of these quantities, virtually no diffusivity data exist except near ambient conditions and for relatively simple systems. Because this study was conducted at moderate pressures and low temperatures, the only option available was to extend the existing information to the conditions of interest. A discussion of this approach follows.

Diffusivities in Binary Gases

The literature contains numerous correlations for predicting binary gas phase diffusivities. Unfortunately, most of these correlations apply to fairly simple systems at ambient temperatures and pressures. Unfortunately, the ranges of temperatures and pressures covered in these correlations do not encompass the conditions of interest in this study. In addition, the presence of high concentrations of hydrogen in the gas phase makes the prediction of binary and multicomponent diffusivities especially difficult.

Because no one correlation appeared to encompass the range of operating conditions studied, several correlations were included in the subroutines DIFFG (SIMPAK) and DIFFGM (MCOMP), with the choice of a particular correlation made in the program input. A discussion of these correlations is presented next.

Chapman-Enskog Theoretical Relationship (1960)

This relationship results from the solution of the Boltzman equation and is often referred to as the Hirschfelder, Bird, and Spotz equation. Assuming that the gas phase can be considered ideal, the binary gas diffusivity can be described by the equation

$$D_{AB} = 0.001858\ T^{3/2}\ [(M_A+M_B)/(M_A M_B)]^{1/2}/[P*SIGAB^2*OMEGA] \qquad (47)$$

where D_{AB} = diffusivity in cm^2/sec
T = temperature, Kelvin
P = pressure, atmospheres
SIGAB = characteristic length, angstroms = (SIGA + SIGB)/2.0
SIGA = radius of molecule A
SIGB = radius of molecule B
OMEGA = diffusion collision integral = f(T,EAB)
EAB = $(EA * EB)^{1/2}$
EA = energy of molecular attraction
EB = energy of molecular attraction

An intermolecular force law must be selected and the values of SIGAB and OMEGA evaluated in order to use this correlation. Values of SIGA (and SIGB) and EA (and EB) from the same source will generally provide consistent results for the evaluation of the collision integral. In this case, the values of SIGA (and SIGB) and EA (and EB) were taken from the data of Svehla (1962) and the diffusion collision integral was evaluated with the expression suggested by Neufeld et al (1972). The Chapman-Enskog relationship is particularly suited for pairs of non-polar molecules.

Wilke-Lee Modification to Chapman-Enskog (1955)

Wilke and Lee suggested an empirical modification to the Chapman-Enskog expression that was obtained by applying this relationship to a large set of diffusion data and statistically analyzing the errors between predicted results and experimental data. Their results gave

$$B = 10.7 - 2.46\ [(M_A + M_B)/(M_A * M_B)]^{1/2} \tag{48}$$

$$D_{AB} = B\ T^{3/2}\ [(M_A + M_B)/(M_A * M_B)]^{1/2}/\{P * \mathrm{SIGAB}^2 * \mathrm{OMEGA}\} \tag{49}$$

This empirical expression represents an improvement over the Chapman-Enskog relationship for many binary systems.

Brokaw Modification to Chapman-Enskog (1969)

For systems containing two polar gases or a polar and non-polar gas, the modification suggested by Brokaw may provide additional accuracy. The effects of molecular polarity is related to the dipole moment in the expression

$$D_{AB} = 0.001858\ T^{3/2}\ [(M_A + M_B)/(M_A * M_B)]^{1/2}/[P\ \mathrm{SIGAB}^2\ \mathrm{OMEGA}'] \tag{50}$$

where $\mathrm{OMEGA}' = \mathrm{OMEGA} + 0.19\ \mathrm{DELAB}^2/(kT/\mathrm{EAB})$
$\mathrm{DELAB} = \mathrm{DELA} * \mathrm{DELB}$
$\mathrm{DELA} = 1.94\mathrm{E}3 * \mathrm{DIPA}^2\ /\{\mathrm{VA}_B * \mathrm{TA}_B\}$
$\mathrm{DELB} = 1.94\mathrm{E}3 * \mathrm{DIPB}^2\ /\{\mathrm{VB}_B * \mathrm{TB}_B\}$
DIPA = dipole moment of A in debyes
DIPB = dipole moment of B in debyes
VA_B = liquid molar volume of A at TA_B
VB_B = liquid molar volume of B at TB_B
TA_B = normal boiling point of A
TB_B = normal boiling point of B

For the degenerate case of zero dipole moment, the expression for the collision integral reduces to the Lennard-Jones relation.

Method of Fuller, Shettler and Giddings (1966)

A somewhat different approach has been reported by Shettler et al that incorporates the concept of atomic diffusion volume of each constituent into the correlation

$$D_{AB} = \frac{0.001 * T^{1.75} * [(M_A + M_B)/(M_A * M_B)]^{1/2}}{P * [(\Sigma v_A)^{1/3} + (\Sigma v_B)^{1/3}]^2} \qquad (51)$$

where v_A, v_B = the atomic diffusion volumes of A and B

This expression has been shown to fit 340 data points well, although most of the data were obtained at room temperature. The atomic diffusion volumes were computed from information provided by Shettler et al (1966).

Although the above correlations were obtained by fitting data at ambient temperatures and pressures, extension to the ranges studied here should not be difficult. Gas phase diffusion coefficients are inversely proportional to pressure, at least at moderate pressures (Reid et al,1977). The influences of temperature and the presence of other components are not well documented and it was assumed that these effects are either accounted for in the correlations or not important.

DIFFUSIVITIES IN MULTICOMPONENT GASES

To determine mixture diffusivities, the expression of Bird et al (1960) was used. This expression, which is based on the Stefan-Maxwell equation, is not limited to the case of a single component diffusing through a stagnant mixture of the other components. To use this expression, an estimate of the binary diffusivities as well as the molar fluxes of each component must be obtained first.

$$\{1.0/D_{i,mix}\} = \sum_{j=1}^{n} \frac{\{1.0/D_{ij}\} * (y_j R_i - y_i R_j)}{R_i - y_i \sum_{j=1}^{n} R_j} \qquad (52)$$

where $D_{i,mix}$ = effective diffusivity of i in j components

Diffusivities from these expressions were used in the suggested correlations for gas phase mass transfer coefficients.

DIFFUSIVITIES IN BINARY LIQUIDS

Because of the difficulty of describing liquids, there exists no single correlation that accurately predicts binary liquid diffusivities. Instead, a variety of correlations have been proposed to account for intermolecular forces, and the shape and size of molecules as factors in the diffusional process. The following paragraph taken from Cussler (1976) puts the situation in perspective:

> "In general, however, diffusion coeficients in many common situations fall in a narrow range around about 0.7E-5 cm^2/sec. In my own laboratory, this narrow range has prompted the generalization known as Toor's law, that "all diffusion coefficients in liquids are equal." ...Theoretical estimation of diffusion in liquids is much less accurate than those for gases. In general, they are useful only for molecules of moderate molecular weight, for which an average accuracy of 20% is commonly claimed. This accuracy would be much more impressive if the diffusion coefficients of the smaller molecules didn't almost all fall in a narrow range around 0.7E-5 cm^2/sec."

In essence, while it is possible that the correlation used for determining a binary liquid diffusivity may be inaccurate, the magnitude of the error may be small if Cussler's observation is correct.

In the program input, one of two binary liquid diffusivity correlations may be chosen for use in subroutines DIFFL (SIMPAK) and DIFFLM (MCOMP). A brief discussion of each follows:

Wilke-Chang Correlation (1955)

This correlation is based on the Stokes-Einstein equation

$$D_{AB} = kT/\ 6\pi\mu_B r_A \qquad (53)$$

where D_{AB} = diffusion coefficient of A in B
k = Boltzman constant
T = absolute temperature
μ_B = viscosity of solvent B
r_A = radius of the solute A particle

Wilke and Chang modified this expression to

$$D_{AB} = \frac{7.4E-8 * \{ X_B M_B \}^{1/2} * T}{\mu_B * V_A^{0.6}} \qquad (54) \qquad\qquad (54)$$

where X_B = association factor for the solvent = 1.9 for methanol

M_B = molecular weight of B

V_A = molar volume of solute A

This correlation has been tested for 251 data points with a reported accuracy of 10%. It should be noted that Davies et al (1967) found that for CO_2 in various solvents, $D_{AB} * \mu_B^{0.45}$ was constant for viscosities from 1 to 27 centipoise.

Scheibel Correlation (1971)

This particular correlation is recommended by Reid et al (1977) for solutes diffusing through organic solvents, although the data used in developing the correlation was in the 0-100°C range. The correlation takes the form

$$D_{AB} = KT/(\mu_B V_A^{1/3}) \qquad (55)$$

where $K = 8.2E-8 [1 + (3V_B/V_A)]^{2/3}$
V_A = molar volume of A
V_B = molar volume of B
$K = 25.2E-8$ if $V_A < V_B$
$= 17.5E-8$ if $V_A < 2.5V_B$

Effects of compositions on binary diffusivity were ignored.

Comparison of Binary Diffusivity Correlations

Before discussing methods for determining mixture diffusivities, some comments should be made on the existing correlations for binary cases. The Wilke-Chang correlation, which is probably the one most widely used in engineering calculations, is best for mixtures where moderate solute-solvent interactions are present. Perhaps its greatest weakness is the fact that the association parameter, X_B, must be determined experimentally. While Wilke and Chang recommend using $X_B = 1.9$ for methanol, Lusis and Ratcliff (1971) suggest using $X_B = 3.3$ because of their success in improving the accuracy of the correlation for systems where hydrogen bonding is a factor. They point out that the larger value of X_B stems from the relatively strong hydrogen bonding potential between the solute and solvent hydroxyl groups. They report that for the diffusion of inert solutes in methanol, using an X_B value of 3.3 improves the average error to 10% from greater than 20% for data taken in the neighborhood of 25°C.

Other researchers have attempted to improve upon Wilke and Chang's correlation. Akgerman and Gainer (1972) developed a correlation based on absolute rate theory that performed better than the Wilke-Chang correlation for small molecules such as H_2 and He, and for solvent viscosities in the 3 to 5 cp range for data collected at temperatures greater than 10°C. Hayduk and Buckley (1972) hypothesized that the diffusivity depends in part on the shape of the molecule. They pointed out that for linear molecules, the binary diffusivity is more than 30% greater than for spherical molecules having the same molar volume at the normal boiling point.

While diffusivity data for various solutes in methanol is virtually nonexistent, some information is available in Reid et al (1977) for the diffusivity of CO_2 in ethanol at 17°C. For the reported value of CO_2 diffusivity in ethanol, given as 3.2E-5 cm^2/sec, the following is noted:

Correlation	% Difference
Wilke-Chang	-48
Scheibel	-62
Lusis and Ratcliff	-49

This accuracy breeds little confidence when considering the need for data describing the diffusion of solutes in methanol at temperatures as low as -40°C. It appears that with the little information available the diffusivities of interest in the system may be at best order of magnitude estimates.

DIFFUSIVITIES IN MULTICOMPONENT LIQUIDS

Given the relative accuracy of the binary diffusivity predictions for CO_2 in ethanol, extension of this information for use in expressions for mixture diffusivities was done reluctantly. Feintuch (1973) presents an interesting discussion of several possible approaches that have been suggested in the literature. He concluded that the necessity of having to execute an engineering calculation makes the use of several of the more theoretical approaches impractical. Instead, he recommended using either the approach previously used by Bird et al for gas mixtures or the modification of the method of Wilke-Chang by Holmes et al (1955).

The method of Bird et al (1960) has been described in the section on gas phase diffusivities. Holmes et al (1962) proposed using a modified molecular weight in the Wilke-Chang correlation that accounts for the presence of other solutes. Their approach was to consider the diffusion of any one solute into the "solvent" as if the system could be treated as a "pseudo-binary". Each solute can be treated in turn with the other solutes factored into the expression through a modified molecular weight expression.

The approaches of Bird et al (1960) and Holmes (1962) et al were included in the model in subroutine MLTDIF (MCOMP); the choice of using either was made in the program input. Because only a single solute transfers in the situation treated by SIMPAK, no mixture diffusivity in the liquid was necessary and the binary coefficient was used.

MASS TRANSFER COEFFICIENTS AND INTERFACIAL AREA

Description of the performance of a packed column requires both transfer coefficients and interfacial areas for mass and heat transfer. These quantities, along with information concerning column hydraulics, will determine packing effectiveness. Although the literature contains a great deal of information about the performance of certain packings with particular systems, prediction of the performance of packings for which data are not available is difficult at best.

Mass transfer data, obtained from a particular column and system, are often reported as the product of the transfer coefficient and the interfacial area. This is because it is often difficult, if not impossible to separate these two quantities. However, using data obtained on one system to predict mass transfer rates in another often requires estimation of both of these quantities. A discussion of predictive methods for mass transfer coefficients and interfacial areas will be presented after a brief review of background information.

MASS TRANSFER COEFFICIENTS

While many investigators have studied the effectiveness of various packings, few have tried to generalize their results to other operating conditions and systems. For this reason, design methods for packed columns still rest to a large extent on experience and vendor advice. Large safety factors have to be used to ensure that the column will meet design expectations (Bolles and Fair, April 1981).

To be able to characterize column performance accurately, mass transfer coefficients in the liquid and gas phases must be known. If this information is not available for the system being used, correlations based on existing experimental data can be used to predict these quantities, (Bolles and Fair, April 1981; Shulman et al, 1955; Onda et al 1968). The accuracy of these correlations depends upon the accuracy of the data used in their development. This is an especially important consideration since experimental difficulties make the measurement of mass transfer coefficients susceptible to significant error.

Liquid-phase mass transfer coefficients are relatively easy to determine because of the existence of several solute-solvent systems where the gas-phase resistance to mass transfer is negligible. Gas phase mass transfer coefficients, on the other hand, are much more difficult to determine because the liquid-phase resistance to mass transfer is rarely negligible. In addition, test systems used to determine gas phase mass transfer coefficients usually involve the complication of thermal effects associated with heats of vaporization, heats of reaction, or heats of solution. These may cause local temperature gradients that result in local changes in phase equilibria as well as changes in surface tension that can affect the film flow on the packing surface; that is, conditions in the bulk phase may not reflect the situation locally.

Several techniques have been used to determine gas phase mass transfer coefficients. A pure liquid can be evaporated into a gas stream or a solute gas can be absorbed into a liquid where the equilibrium partial pressure is zero because of an irreversible chemical reaction. Both techniques suffer from the possible influence of heat effects but give reasonably good results (Shulman et al, 1955; Sridharan and Sharma, 1976; Alper et al, 1980). Another approach includes accounting for the liquid-phase resistance in determining the gas phase transfer coefficient. Unfortunately, while this method is feasible, few systems are sufficiently characterized for this approach to be accurate. Most of the work done to determine gas-phase transfer coefficients is carried out by evaporating liquids from a known contacting surface area which has been saturated with the liquid.

Some of the difficulties associated with determining gas-phase mass transfer coefficients arise from the fact that the true gas-phase coefficients are generally independent of liquid flow rate. This is true over a wide range of superficial gas velocities. Some researchers mistakenly associate the effect of a changing liquid flow rate with the coefficient when the effect can be entirely accounted for by the changing interfacial area (Sahay and Sharma, 1973).

Even with these experimental difficulties, there are methods for predicting the mass transfer coefficients in both the gas and liquid phases. Four of these methods have been used in this study and are described later.

INTERFACIAL AREAS

While it is relatively easy to determine the dry surface area associated with a particular type and size of packing, it is difficult to determine the amount of the packing surface that is effectively used in an irrigated section of packing. Several factors are important: The type of process for which the column is used will influence the amount of surface area; the surface tension of the liquid passing through the packing determines the wetting characteristics; and the shape, size, and material used for packing are also important considerations.

Most researchers who propose a particular correlation for a mass transfer coefficient also suggest an expression for the interfacial area. In effect, by requiring that their mass transfer coefficient correlation be used with a particular expression for interfacial area, they provide a single correlation for the product of the coefficient and the interfacial area.

Although there are discrepancies in the results various researchers obtain for the interfacial area, there is agreement on the factors that determine this quantity. Shulman et al (1955) postulated that the liquid hold-up in a packing is a combination of the static hold-up and the dynamic hold-up

$$h_{tot} = h_{static} + h_{dynamic} \tag{56}$$

The static hold-up is defined as the amount of liquid that will remain in the column packing after the liquid feed to the column is stopped and the column is allowed to drain. In effect, it results from a balance between the surface tension forces that hold the liquid in the packing and the force of gravity which tends to drain the liquid from the bed. The dynamic hold-up, defined as the difference between the total and static hold-up, directly affects the mass and heat transfer rates since it determines the residence time of the liquid in the packing. Both the static and dynamic hold-up are important factors in determining the amount of effective surface area in a particular packing.

Shulman et al (1955) made a distinction between effective interfacial area and wetted area. They pointed out that for certain situations, especially physical absorption, the entire wetted surface of the packing is not involved in the mass transfer process. Their approach to correlating mass transfer coefficients and interfacial areas included the definition of an effective interfacial area that is less than or equal to the wetted surface area. The wetted area generally increases as the packing size decreases but the effective interfacial area is smallest for the smallest packing. Shulman et al (1955) reason that small packings have large static hold-ups which are found in the packing as stagnant pools. Most of the large wetted area thus becomes ineffective, especially during physical absorption processes, because these pools of liquid quickly reach equilibrium. On the other hand, larger packings have correspondingly smaller static hold-up allowing the liquid to run through the packing more easily. Here the effective area approaches the wetted area.

More recently, Puranik and Vogelpohl (1974) conducted an interesting study of effective interfacial areas in packed columns operated below the loading point. Their development of a generalized correlation for effective interfacial area took into account several different mass transfer processes: physical absorption, chemical absorption and vaporization. Following the approach of Shulman et al (1955)and Elgin et al (1943), they assumed that the entire static hold-up, h_s, correlates with the static interfacial area, and the dynamic hold-up correlates with the dynamic interfacial area. The total wetted surface area, which is also the maximum area available for mass transfer, can be expressed as:

$$a_{wet} = a_{max} = a_{static} + a_{dynamic} \tag{57}$$

For the cases of vaporization and chemical absorption, effective interfacial area closely approximates wetted area. Using data available in the literature, Puranik and Vogelpohl correlated wetted surface area with a stated accuracy of 20% by the expression

$$a_w/a_t = 1.05(Re)^{0.047} (We)^{0.135}(\sigma / \sigma_c)^{-0.206} \tag{58}$$

where a_w = wetted surface area
a_t = total surface area
Re = Reynolds number = $L'/(a_{tot\ L})$
We = Weber number = $L'^2/(a_{tot\ L}\ \ )$
L' = superficial liquid rate
σ = surface tension of liquid
σ_c = surface tension of packing surface

To determine effective interfacial area for physical absorption, they first calculated static area

$$a_{static}/a_{tot} = 0.229 - 0.091 \ln(We/Fr) \tag{59}$$

where Fr = Froude number = $L'^2 a_{tot}/\rho_L{}^2 g$
and $We/Fr < 12.5$

The effective interfacial area for physical absorption can then be determined as

$$(a_{physical}/a_{tot}) = (a_{wet}/a_{tot}) - (a_{static}/a_{tot}) \tag{60}$$

Note that for cases where the term [We/Fr] is greater than 12.5, the area for physical absorption is the same as the wetted area.

The work reported by Puranik and Vogelpohl (1974) is interesting because they attempt to quantify the difference between wetted surface and effective interfacial areas on some physical basis. For cases involving vaporization of the liquid, it is expected that both static and dynamic portions of the wetted area are involved in the mass transfer process. For chemical absorption cases, all of the dynamic area would be expected to be important as well as some portion of the static area depending on the capacity of the static area to react with and absorb the solute. As the concentration of the active component in the chemical solvent is increased, the effectiveness of the static area would be expected to increase. In physical absorption cases, the pools of semi-stagnant liquid that comprise the static area will quickly become saturated with solute and rendered ineffective in the mass transfer process. In this instance only the dynamic area of the packing associated with the liquid moving through the packing should be considered effective.

Several researchers including Onda et al (1968) assume that the entire wetted surface should be considered in any situation. Others including Fair et al (April 1981) avoid separating interfacial area and mass transfer coefficient and correlate the product of the two. A discussion of the approaches of Shulman et al (1955), Onda et al (1968) and Fair et al (April 1981) follows. These three correlations are factored into the model as well as that of Puranik and Vogelpohl (1974), which combines an effective interfacial area correlation with Shulman's mass transfer coefficient relationship. The option chosen must be specified in the program input.

CORRELATIONS USED IN MODEL

Because there is no widely accepted correlation for either mass transfer coefficients or interfacial area in packed columns, four different options have been included in the computer programs SIMPAK and MCOMP. Each of these four options will be discussed in this section. The option to be used in a particular computation is chosen in the program input.

Relationship of Shulman et al (1955)

In a series of articles appearing in the literature from 1955 to 1971, Shulman and co-workers considered several aspects of packed column operation. Their approach was to separate mass transfer coefficients from corresponding interfacial areas and to correlate each independently. Experimental data were provided from results obtained by sublimation of napthalene into a flowing gas. Since the surface area of the solid napthalene was known, interfacial area-free mass transfer coefficients could be evaluated from the data. Liquid-phase mass transfer coefficients were determined by measuring $k_L a$ and dividing through by the interfacial area previously determined.

Correlations of the mass transfer coefficients, take the following form:

$$F_{G_j} = 1.195G[d_s\, G'/\mu_G(1 - EPS_o)]^{-0.36} Sc_{G_j}^{-2/3} \tag{61}$$

$$F_{L_j} = (25.1D_{L_j}/d_s MW_L)[d_s L'/\mu_L]^{0.45} Sc_{Lj}^{0.5} \tag{62}$$

where F_G, F_L = Drew-Colburn mass transfer coefficients

G, L = superficial velocities (molar)
G', L' = superficial velocities (mass)
μ_G, μ_L = viscosity
ρ_G, ρ_L = density
MW_L = average molecular weight of liquid phase
Sc_{L_j} = Schmidt number for j in liquid = $\mu_L / \rho_L D_{L_j}$
Sc_{G_j} = Schmidt number for j in gas = $\mu_G / \rho_G D_{G_j}$
D_{L_j} = diffusivity of j in liquid
D_{G_j} = diffusivity of j in gas
d_s = diameter of sphere having surface area of particle
EPS_o = operating void space in packing = $EPS - h_{tot}$
EPS^o = total void space in dry packing
h_{tot} = total liquid hold-up

The interfacial area is reported by Shulman et al in an extensive series of graphs that have been represented numerically by Treybal (1980). The interfacial area for non-aqueous systems can be determined by multiplying the interfacial area for the aqueous case by the ratio of the hold-up of the aqueous case to the non-aqueous case

$$a_e = a_{water}(h_{tot}/h_{w,tot}) \tag{63}$$

where a_e = effective interfacial area
a_{water} = effective interfacial area for aqueous systems
h_{tot} = total hold-up
$h_{w,tot}$ = total hold-up for aqueous systems

In the current study, where 1/4" ceramic Intalox saddles were used, a modification of the Shulman relationship for Berl saddles was used. First, Intalox saddles were assumed to be sufficiently similar in shape to Berl saddles in that they have the same performance characteristics. Additionally, since 1/2" Berl saddles were the smallest packing size studied by Shulman, it was necessary to assume that, with the exception of the value for d_s, the same relationships that apply to the 1/2" packing could be used for 1/4" packing. The following procedure was used to calculate the interfacial area and mass transfer coefficients in the model:

1. Compute the physical and transport properties at the average of the temperatures of the bulk flow and the interface.

2. Compute the static, total, and dynamic hold-ups for the aqueous case using the relationships for 1/2" Berl saddles with the value of d_s of 1/4" Intalox saddle packing (d_s = 0.027708 feet).

3. Determine the value of the correction factor for non-aqueous case and compute the corrected total liquid hold-up.

4. The mass transfer coefficients are then computed using the value of d_s for 1/4" Intalox saddle packing and used with the interfacial area value in the model.

Heat transfer coefficients are also needed in the model. These are determined using the j-factor analogy for heat and mass transfer assuming the same interfacial area applies for both phenomena.

$$h_G = \left[\frac{1.195*C_G*G'}{Pr^{2/3}}\right] \left[\frac{d_s * G'}{\mu_G*[1 - EPS_o]}\right]^{-0.36} \tag{64}$$

$$h_L = \left[\frac{25.1*k'_L}{d_s}\right] \left[\frac{d_s*L'}{\mu_L}\right]^{0.45} Pr_L^{0.45} \tag{65}$$

where C_G = heat capacity of gas phase

Pr_G = Prandtl number in gas phase = $C_G\mu_G/k'_G$

Pr_L = Prandtl number in liquid phase = $C_L\mu_L/k'_L$

k'_G, k'_L = thermal conductivity

The Ackerman (1937) correction factor, $h_G'a$, is used for the gas phase to account for concentration gradients. The correction is made as follows:

$$h_G'a = \frac{\Sigma C_{Gj}\, dG_j/dZ}{1 - EXP\{[\Sigma C_{Gj}G_j\, dG_j/dZ]/h_G a\}} \tag{66}$$

No correction was used for the liquid phase.

Relationship of Onda et al (1968)

A more recently developed correlation was published by Onda et al (1968). This correlation resulted from the statistical analysis of data available in the literature. Data reported as the product of the mass transfer coefficient and the interfacial area were divided by the wetted area as determined by Onda et al.

$$a_{wet}/a_{tot} = 1.0 - \exp\{-1.45[\sigma_c/\sigma]^{0.75} Re_L^{0.1} Fr^{-0.05} We^{0.2}\} \qquad (67)$$

where the terms are as previously defined.

The resulting expressions for mass transfer coefficients include consideration of data for water and organic solvents. Accuracies of 20% are reported. The liquid phase transfer coefficient, expressed in the form of the Drew-Colburn coefficient, is

$$F_{Lj} = 0.0051(\rho_L/\mu_L g)^{-1/3} Re_L^{2/3} Sc_{L_j}^{-0.5} (a_{tot} d_p)^{0.4} \rho_L/M_L \qquad (68)$$

where Re_L = Reynolds number in the liquid = $L'/(a_{wet}\mu_L)$

and d_p = nominal diameter of packing

For packing less than 1/2" in size, a modified gas phase coefficient is suggested

$$F_{Gj} = 2.00\ P\ \{a_{tot} D_{Gj}/RT\}\ Re_G^{0.7}\ Sc^{1/3} a_{tot} d_P \qquad (69)$$

where P = total pressure
Re_G = Reynolds number in the gas phase = $G'/a_{tot}\mu_G$

It is interesting to note that the constant 2.00 replaces 5.23 which was used to correlate data for larger packings.

The approach used by Onda et al (1968) calls for use of the wetted surface area with the mass transfer coefficient. Their recommendation is correlation of data for both absorption and vaporization with the wetted area. This is different from the approach suggested by other workers (Onda et al, 1968; Sridharan and Sharma, 1976) and the reported accuracy of 30% may account for the failure to note the distinction.

The heat transfer coefficients are calculated through the j-factor analogy:

$$h_G = 2.00k'_G a_{tot} Re^{0.7} (a_{tot} d_p)^{-2.0} Pr_G^{1/3} \tag{70}$$

$$h_L = 0.0051 \{\rho_L/\mu_L g\} C_L \rho_L Re_L^{2/3} Pr_L^{-0.5} (a_{tot} d_p)^{0.4} \tag{71}$$

The Ackerman (1937) correction was again used to modify the gas phase heat transfer relationship as was done with the Shulman's correlation. The wetted surface area is assumed to be identical to the interfacial area for mass and heat transfer.

Correlation of Fair et al (1981, 1960)

Yet another relationship for packed column mass transfer coefficients and interfacial areas was developed by Fair and co-workers (1960a, 1960b) and has since been updated to include more recent data (April 1981). This approach was to correlate heights of mass transfer units for gas and liquid phases. Heights of mass transfer units can be defined in the form of the Drew-Colburn coefficients F_G and F_L, and active area for mass transfer AA.

$$HTU_G = G(\text{MOLAR FLOW RATE})/F_G AA \tag{72}$$

$$HTU_L = L(\text{MOLAR FLOW RATE})/F_L AA \tag{73}$$

The product of the mass transfer coefficient and the interfacial area can then be found once the height of a transfer unit is known.

The Fair correlation applies only to packing sizes of at least 1/2" and to column diameters of at least 0.6 feet. Since Fair correlated much of the same data as Onda (1968) who considered smaller packing and neglected the influence of column diameter, it is conceivable that the Fair's correlation may be applicable to smaller packing sizes and column diameters. With this in mind, the correlation was programmed into the computer model as transfer coefficient option 3.

Fair correlated the height of a gas phase transfer unit as

$$HTU_G = \psi (d_c')^m (Z_p/10)^{1/3} (Sc_G)^{1/2} \{G' f_\mu f_\rho f_\sigma\}^{-n} \tag{74}$$

where HTU_G = height of a gas phase transfer unit, feet

ψ = packing parameter, feet
= f(packing type, size, % of flooding velocity)
G' = gas mass velocity, lb/ft^2-hr
d_c' = the minimum of the column diameter or 2.0 feet
m = 1.11 for ceramic saddles
n = 0.5 for ceramic saddles
f_μ = $(\mu_L/\mu_W)^{0.16}$
f_ρ = $(\rho_L/\rho_W)^{-1.25}$
f_σ = $(\sigma_L/\sigma_W)^{-0.8}$
Z_p = height of packed column, feet

The term $(Z_p/10)$ is a correction factor for liquid distribution in the column. In this model, Z_p is initially assigned a value of 2.0 and is replaced by the calculated height of packing at the end of each successive iteration. The parameter, ψ, is a function of the percent of flooding velocity for a given packing size and type. Eckert (1970) used a least squares fit of flooding data to correlate flooding velocities as follows: First, a parameter B is evaluated from liquid and gas flow rates and densities of the liquid and gas.

$$B = (L'/G')(\rho_G/\rho_L)^{1/2} \tag{75}$$

Next, a parameter A is determined from the correlation

$$\ln(A) = -3.92 - 1.09\ln(B) - 0.12\{\ln(B)\}^2 \tag{76}$$

The flooding velocity G'(Flooding) can then be calculated from the definition of A

$$A = \left[\frac{G'(\text{Flooding}) * F_p}{g * \rho_G * \rho_L}\right] \left[\frac{\rho_W}{\rho_L}\right]\left[\frac{\mu_L}{\mu_W}\right]^{0.2} \tag{77}$$

where F_p = packing factor = 725
g = gravitational constant

Once the flooding velocity is determined, the actual gas flow rate can be used to determine the percentage of flooding velocity and, finally, flooding velocity can be calculated and used to estimate the packing parameter :

$$\psi = 0.29 + 2.02(\%\mathrm{Flood}) - 0.03(\%\mathrm{Flood})^2 \qquad (78)$$

$$+ 3.106\mathrm{E}{-4}(\%\mathrm{Flood})^3 - 1.136\mathrm{E}{-6}(\%\mathrm{Flood})^4 \qquad (79)$$

Knowing 1, the value of the gas phase transfer unit can be determined which provides the information to compute the product of the mass transfer coefficient and the interfacial area

$$F_G AA = G(\mathrm{MOLAR})/HTU_G \qquad (80)$$

The liquid phase height of a transfer unit is expressed as

$$HTU_L = \phi / C_{FL} \, (Z_p/10)^{0.15} \, Sc_L^{1/2}$$

where HTU_L = height of a liquid phase transfer unit, feet

ϕ = packing parameter for liquid phase, feet

= f(packing type, size, liquid flow rate)

C_{FL} = correction factor for mass transfer in the liquid phase as a function of flooding velocity

ϕ ranges from 0.03 to 0.15 feet for 1/2" ceramic saddles and is correlated as a function of the liquid flow rate:

$$\begin{aligned} \ln \phi &= 0.084762 \ln L' - 3.7512 && 200 \le L' \le 900 \\ &= 0.283303 \ln L' - 5.1050 && 900 \le L' \le 9000 \\ &= 0.331787 \ln L' - 5.5572 && 9000 \le L' \le 50{,}000 \end{aligned}$$

C_{FL} ranges from 0.45 to 1.00 for 1/2" ceramic saddles and can be expressed as

$$C_{FL} = 1.00 \qquad (\text{if } F_r \neq 0.4375)$$

$$= 6.37 - 37.46F_r + 96.91F_r^{2} - 109.14F_r^{3}$$
$$+ 44.02F_r^{4} \qquad (\text{if } F_r = .4375)$$

where $F_r = G'/G'(\text{Flooding})$ from the Eckert model (1970).

The liquid phase transfer unit can be computed once C_{FL} and ϕ are known. Z_p was treated as in the case of the gas phase height of a transfer unit. Once HTU_L was known, the product of the liquid phase mass transfer unit and the interfacial area could be determined

$$F_L AA = L(MOLAR)/HTU_L \tag{81}$$

Heat transfer coefficients were determined through the j-factor analogy and the height of a heat transfer unit

$$HTU_G(\text{heat transfer}) = C_G \; G'/(h_G AA) \tag{82}$$

$$HTU_L(\text{heat transfer}) = C_L \; L'/(h_L AA) \tag{83}$$

The transfer units are found by replacing the Schmidt number by the Prandtl number in equations (74) and (80). The correction suggested by Ackerman (1937) is again applied to the gas phase heat transfer unit.

The assumption made in using the Fair correlation was that the relationship for 1/2" saddle packing could be used for 1/4" packing. Inspection of the differences between 1" and 1/2" packing parameters showed that this assumption was plausible.

Combination of Shulman (1955) and Puranik and Vogelpohl (1974)

Because the Shulman correlations do not include data for packing sizes less than 1/2", it was expected that application of these correlations to the packing used in this work may lead to error. However, since only the packing size is included in the mass transfer coefficient correlations, it is reasonable to expect that these correlations might be accurate for 1/4" packing, even though the effective interfacial area relationship suggested by

Shulman may be inaccurate. With this in mind, option four in the program input combines the Shulman mass transfer coefficient correlation with the effective interfacial relationship of Puranik and Vogelpohl.

Comparison of the use of this correlation to those of Shulman, Onda, and Fair is discussed later.

EXPERIMENTAL METHOD

Details of the gasifier and the entire pilot plant are provided in the report described by Ferrell et al (1980), that is volume I of this series. A detailed description of the packed absorption column and its operation will be provided here.

EQUIPMENT

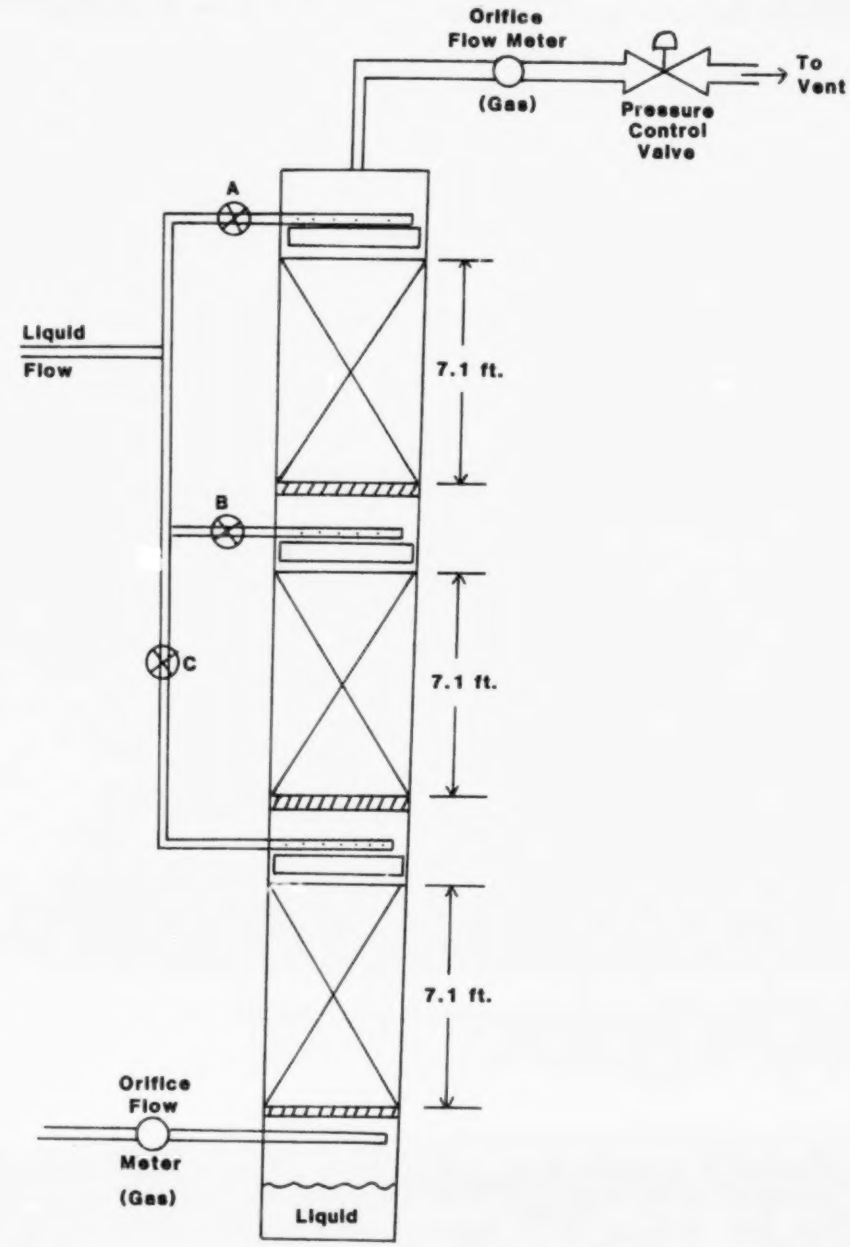
The absorber used in this study was constructed from a length of nominal 5" Schedule 40 stainless steel pipe with an internal cross-sectional area of 0.139 square feet. The column was built with three 7.1 foot sections packed with 1/4" ceramic Intalox saddles. Any or all of these sections could be used in mass transfer studies. Each section contained two flow redistributors to prevent liquid channeling. The column was insulated and approached adiabatic operation. A schematic of the column is shown in Figure 8. Details on the instrumentation, process control and data acquisition used during experimental runs are described by Ferrell et al (March 1980).

DATA ANALYSIS

In developing a mathematical model to describe the packed absorber operation, a test of model predictions against experimental results is desirable. The test depends on how the problem is formulated. If it is a design formulation, then a determination of the height required to effect a specified separation must be determined by both the model and experiment. If it is an analysis formulation, then a comparison of experimental and predicted outlet gas compositions, for a given packing height must be made. In addition, a favorable comparison of liquid and gas compositions at various packing heights with those predicted by the model predictions would support the validity of the model. Another possible basis of comparison is the liquid or gas temperature profile in the column. The applicability of this approach is more appropriate where the exotherm due to absorption is significant; the temperature profiles are indicative of the amount of mass transfer taking place when adiabatic operation prevails or when non-isothermal effects can be accounted for.

Figure 8

Schematic of Pilot Plant Absorption Column


Orifice
Flow Meter
(Gas)
Pressure
Control
Valve
To
Vent
A
Liquid
Flow
7.1 ft.
B
C
7.1 ft.
7.1 ft.
Orifice
Flow
Meter
(Gas)
Liquid

During the early stages of operation, it became apparent that a firm basis for comparison of experimental and predicted results would be difficult to establish. Although the column had been built with two sampling points located in each section, attempts to draw liquid or gaseous samples were unsuccessful. Because the column was operated at pressures in excess of 30 atmospheres and temperatures as low as $-35^{\circ}F$, the task of procuring a sample from within the packing proved to be extremely difficult. Several methods were tried but in almost every case, entrainment of one phase with the other made the information obtained qualitative at best.

As intended during the design of the system, the use of the three sections of packing as an experimental variable was considered as a means of comparing experimental and predicted results. The concept here was to vary the packed height and note the differences in the removal efficiency of the compounds entering the column. Information obtained during the first trial runs using CO_2 as a solute showed that the high solubility of the acid gas in methanol at the conditions at which the system was operated meant that less than one packed section was needed for substantial recoveries of the solute. System conditions were varied over the available ranges, but the results showed that little or no carbon dioxide was present in the exiting gas stream. While it could be concluded that most of the entering CO_2 could be removed using 7.1 feet of packing for almost every realistic combination of process variables, the amount of packing required in each case was not the same. Thus, some means to determine the amount of packing required to remove specified amounts of solute was needed in order to extrapolate pilot plant results to more industrially significant situations.

One of the characteristics of physical absorption systems is the evolution of large amounts of heat upon the solution of large amounts of very soluble gases in the liquid. This is especially true for the absorption of acid gases such as CO_2, H_2S and COS, and hydrocarbons such as benzene and propane. Because the heat released is indicative of the amount of mass transfer taking place, this represents a way in which experimental and predicted results could be compared. This fact had been recognized in the design of the column but the two temperature sensors located in each column section were positioned too far above the bottom of the packing; that is, they did not provide data in the most important part of the column. In fact, early runs with CO_2 as the solute showed that the two temperature sensors in the bottom section of the column gave values close to the inlet liquid temperature while the temperature of the exiting liquid was often more than $40^{\circ}F$ higher than this value.

While it was apparent that temperature sensors in the bottom 2.5 feet of packing would provide useful information, installation of additional sensors was not possible. The column was built with a pressure rating of 540 psig and drilling or machining on the column walls would endanger the integrity of the column. With these limitations in mind, it was decided to install temperature sensors on the column exterior beneath the insulation. Using platinum temperature sensors obtained from Rosemount, Inc., Minneapolis, Minnesota (Model 118MF100A), the temperatures in the bottom part of the column could be monitored.

After some experimentation, an effective method of securing these sensors to the column wall was developed using a silicon conducting paste. By moving the sensors around the circumference of the column at a given height, it was noted that no significant radial temperature gradients existed. It was assumed consequently that these sensors accurately reflected the liquid temperature profile inside the column and thereby provided a basis for comparison of the predicted and experimental results in this study.

Soon after the external temperature sensors were installed, it became apparent that the placement of the gas distributor in the bottom of the column might be responsible for end effects, i.e. absorption of gases below the bottom of the packing. While in an industrial situation the presence of end effects at the bottom of the column may enhance column capacity, attempting to model this situation is a difficult undertaking. The uncertainty in describing the mass transfer action below the bottom of the packing would likely yield questionable simulation results.

To avoid this problem, the column gas distributor was relocated from a point 0.7 feet below the bottom of the packing to a position immediately below the bottom of the packing. This modification prevents any absorption of acid gases (CO_2, H_2S and COS) in the spray of liquid below the bottom of the packing making the modeling approach more straightforward.

EXPERIMENTAL PROCEDURE

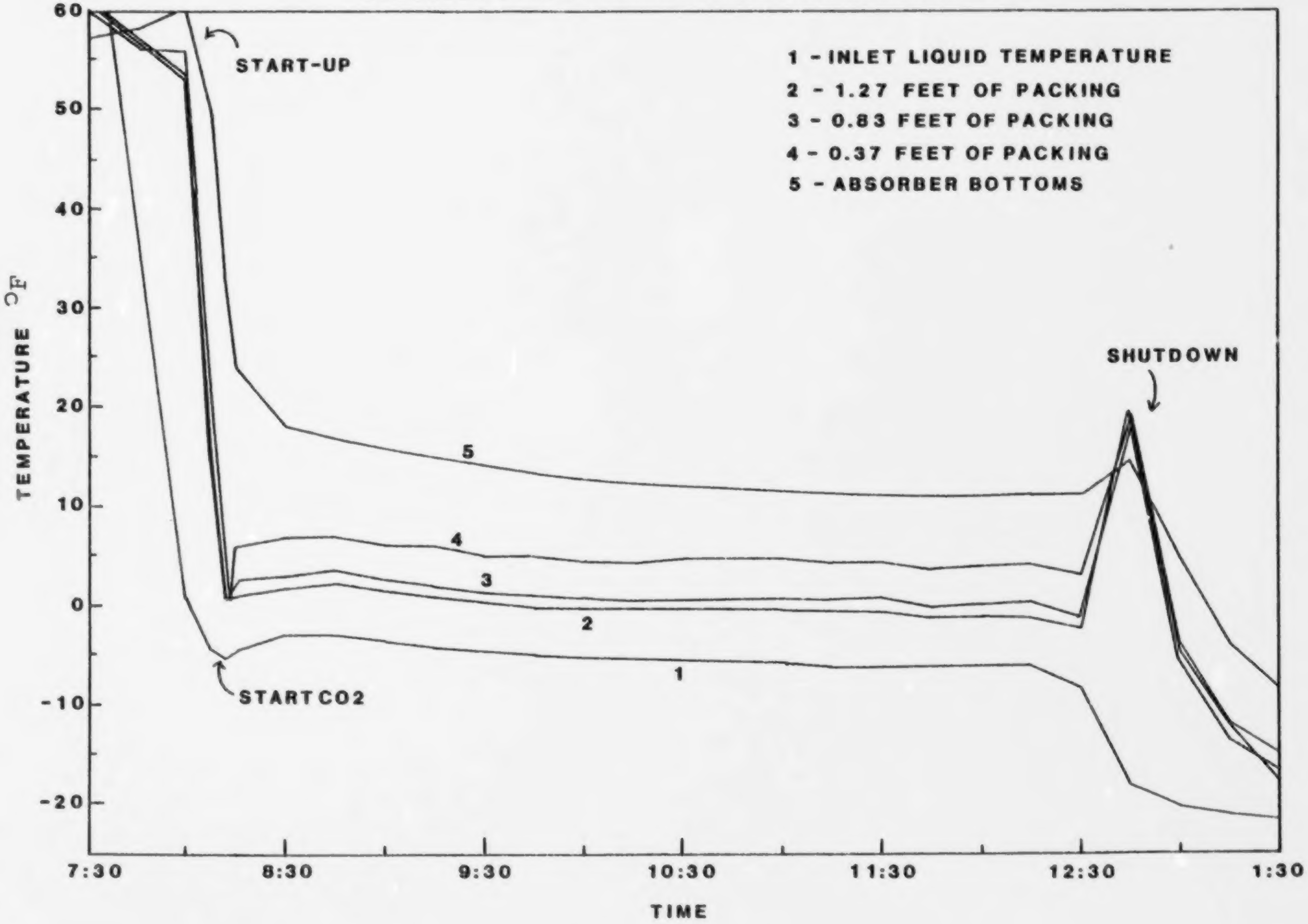
The data required to compare experimental results with model predictions were generated in two ways. Syngas runs involved the use of a gas mixing manifold where gas was fed from bottles and metered to the absorber through a flow controller. Integrated runs involved the use of the fluidized bed gasifier to manufacture gas to be fed to the absorber. The procedures used to operate the absorber and the rest of the acid gas removal system were similar in both types of runs.

Start-up procedures for the absorber are discussed in detail in Part I of this study (Ferrell, et al, 1980). In brief, the column was first pressurized to the specified set point with N_2 while refrigerated methanol was circulated through the system. When the column reached operating pressure and the system had cooled to a point close to the desired operating temperature, either syngas or coal gas was fed to the absorber. The presence of large amounts of CO_2 in this gas immediately became apparent by the temperature rise in the liquid associated with the heat of solution. The entire system, including the flash tank and stripper, was then allowed to come to steady state.

Steady state conditions were characterized by unchanging flow rates, temperatures, pressures, and gas compositions throughout the acid gas system. Figure 9 illustrates the start-up and steady state periods as seen through the wall temperature sensors for syngas run AM-50. During what was defined as steady state, the measured temperatures were essentially constant.

Figure 9

ABSORBER COLUMN TEMPERATURE PROFILE FOR AM-50



The temperature profiles in Figure 9 are representative of both syngas and integrated runs.

After steady state was achieved, gas samples at the inlet to the absorber (sour gas) and the outlets of the absorber (sweet gas), the flash tank (flash gas), and the stripper (acid gas) were taken. After this first sampling period, the system was operated for at least an hour before a second set of samples are taken. After the second set of samples, the system was usually shut down and the run concluded.

After conclusion of the run, collected gas samples were analyzed and used to compute a material balance for the entire system. The methods used to compute this balance are described by Ferrell et al (1980) in Part I of this study. The results from the material balance were then used to assess the quality of the run. Deviations of more than 10% for either the overall balance or the individual component balances usually resulted in disallowing the results of the run. Some judgment was used, however, in analyzing these results, especially if the reason for the discrepancy in the material balance was known. Also, for several components fed to the system at concentrations less than one mole percent, mass balance deviations of greater than 10% may not have resulted in disallowing the run results. As a rule, however, the mass balance results from a particular run met the given criteria. The mass balance results for the both syngas and integrated runs are included in the report by Ferrell et al (1981).

Along with the computation of mass balances, all temperatures, pressures, flow rates, differential pressures, and gas compositions were averaged over the designated steady state period. The computer program, AVERAG, determined averages of all measured variables collected by the data acquisition system. This program, as well as other features of the data acquisition system, are discussed in detail by Willis (1981). These averaged values were used in the data analysis and eventually in the comparison of the experimental data with the model prediction.

DISCUSSION OF RESULTS

This section discusses the results from both syngas and integrated runs and compares these results to model predictions. Although only 21 of the 60 attempted runs are presented, most of the other runs served useful purposes such as providing column hydraulic information or establishing effective operating procedures. A summary of all 60 runs is shown in Table 5.

In the ensuing discussion, the prefix "AM" refers to acid gas removal runs where methanol is used as the solvent, and syngas was used as the feed to the acid gas removal system. The prefix "AMI" refers to integrated runs with methanol as the acid gas removal solvent. For gasifier runs, the prefix "GO" means that the gasifier run was oxygen-blown instead of air-blown. In Table 5, all integrated runs include the number of the corresponding gasifier run.

Many of the AGRS runs made prior to AM-24 were used to develop operating procedures, sampling methods and to check out analytical techniques. Most of the runs made after this point, however, resulted in useful operating information.

SYNGAS RUNS

The following objectives were established for runs made with synthetic gas mixtures of CO_2 and N_2 comprising the feed gas to the absorber:

- Develop and refine operating procedures, sampling techniques, and analytical methods for the acid gas removal system.
- Check out system performance under conditions similar to those used in integrated runs.
- Determine the capability of the wall temperature sensors installed to monitor the liquid temperature profiles in the column.
- Check to see if instrument calibrations, sampling and analytical techniques resulted in closed material balances.
- Provide information necessary to compare model prediction with experimental results.

TABLE 5
AGRS RUN SUMMARY

RUN	DATE	TYPE	PURPOSE	RESULT
1	02/09/79	SYNGAS	TEST	SUCCESSFUL
2	02/25/79	SYNGAS	TEST	SUCCESSFUL
3	03/08/79	SYNGAS	HYDRAULIC DATA	SUCCESSFUL
4	03/14/79	SYNGAS	TEST	SUCCESSFUL
5	05/17/79	SYNGAS	HYDRAULIC DATA	SUCCESSFUL
6	06/05/79	SYNGAS	MASS TRANSFER	SUCCESSFUL
7	06/05/79	SYNGAS	MASS TRANSFER	SUCCESSFUL
8	07/03/79	SYNGAS	MASS TRANSFER	SUCCESSFUL
9	07/05/79	SYNGAS	MASS TRANSFER	POOR MASS BALANCE
10	07/12/79	SYNGAS	TEST LIQ. SAMPLES	POOR MASS BALANCE
11	07/17/79	SYNGAS	TEST LIQ. SAMPLES	SUCCESSFUL
12	07/24/79	INT.(GO-25)	MASS TRANSFER	MECH. PROBLEMS
13	08/14/79	SYNGAS	MASS TRANSFER	SUCCESSFUL
14	08/29/79	INT.(GO-28)	MASS TRANSFER	POOR MASS BALANCE
15	09/12/79	INT.(GO-29)	MASS TRANSFER	POOR MASS BALANCE
16	10/11/79	INT.(GO-30)	MASS TRANSFER	SUCCESSFUL
17	10/24/79	SYNGAS	MASS TRANSFER	SUCCESSFUL
18	11/01/79	SYNGAS	MASS TRANSFER	SUCCESSFUL
19	11/07/79	SYNGAS	TUNE CONTROL LOOPS	SUCCESSFUL
20	11/14/79	SYNGAS	TUNE CONTROL LOOPS	SUCCESSFUL
21	11/29/79	SYNGAS	MASS TRANSFER	POOR MASS BALANCE
22	02/19/80	INT.(GO-38)	MASS TRANSFER	POOR MASS BALANCE
23	02/26/80	INT.(GO-47)	MASS TRANSFER	INSTRUMENT PROB.
24*	03/06/80	INT.(GO-48)	MASS TRANSFER	SUCCESSFUL
25*	03/26/80	SYNGAS	MASS TRANSFER	SUCCESSFUL
26*	04/01/80	SYNGAS	MASS TRANSFER	SUCCESSFUL
27*	04/15/80	SYNGAS	MASS TRANSFER	SUCCESSFUL
28	04/17/80	SYNGAS	MASS TRANSFER	POOR MASS BALANCE
29	05/13/80	INT.(GO-53)	MASS TRANSFER	POOR MASS BALANCE
30*	05/27/80	INT.(GO-54)	MASS TRANSFER	SUCCESSFUL
31	06/05/80	INT.(GO-56)	MASS TRANSFER	MECH. PROBLEM
32*	06/12/80	INT.(GO-57)	MASS TRANSFER	SUCCESSFUL
33*	06/19/80	SYNGAS	MASS TRANSFER	SUCCESSFUL
34*	06/24/80	SYNGAS	MASS TRANSFER	SUCCESSFUL
35*	06/26/80	SYNGAS	MASS TRANSFER	SUCCESSFUL
36*	07/18/80	INT.(GO-59)	MASS TRANSFER	SUCCESSFUL
37*	07/25/80	INT.(GO-60)	MASS TRANSFER	SUCCESSFUL
38*	09/10/80	INT.(GO-61)	MASS TRANSFER	SUCCESSFUL
39	09/16/80	SYNGAS	MASS TRANSFER	SUCCESSFUL
40	11/05/80	SYNGAS	HYDRAULIC DATA	GASIFIER PROBLEMS
41	11/06/80	INT.(GO-67B)	MASS TRANSFER	SUCCESSFUL
42	11/12/80	SYNGAS	HYDRAULIC DATA	GASIFIER PROBLEMS
43*	11/19/80	INT.(GO-68)	MASS TRANSFER	SUCCESSFUL

RUN	DATE	TYPE	PURPOSE	RESULT
44*	12/15/80	INT.(GO-69B)	MASS TRANSFER	SUCCESSFUL
45*	01/08/81	INT.(GO-70)	MASS TRANSFER	SUCCESSFUL
46	01/21/81	INT.(GO-71)	MASS TRANSFER	GASIFIER PROBLEMS
47*	01/27/81	INT.(GO-71B)	MASS TRANSFER	SUCCESSFUL
48	02/08/81	SYNGAS	HYDRAULIC DATA	SUCCESSFUL
49	02/10/81	SYNGAS	HYDRAULIC DATA	SUCCESSFUL
50*	02/12/81	SYNGAS	MASS TRANSFER	SUCCESSFUL
51	02/17/81	SYNGAS	HYDRAULIC DATA	SUCCESSFUL
52*	02/19/81	INT.(GO-72)	MASS TRANSFER	SUCCESSFUL
53*	03/12/81	INT.(GO-73B)	MASS TRANSFER	SUCCESSFUL
54	03/25/81	INT.(GO-74)	MASS TRANSFER	GASIFIER PROBLEMS
55*	04/14/81	SYNGAS	MASS TRANSFER	SUCCESSFUL
56	04/23/81	SYNGAS	MASS TRANSFER	SAMPLING ERROR
57*	04/28/81	INT.(GO-76)	MASS TRANSFER	SUCCESSFUL
58	05/07/81	INT.(GO-77)	MASS TRANSFER	GASIFIER PROBLEMS
59*	05/20/81	INT.(GO-78)	MASS TRANSFER	SUCCESSFUL
60*	06/10/81	INT.(GO-79)	MASS TRANSFER	SUCCESSFUL

All of the above objectives were met in the series of syngas runs.

The syngas runs made for the purpose of obtaining mass transfer information were made using a feed gas mixture of CO_2 and N_2. Initially, plans included using mixtures of other gases, particularly H_2S and COS, but for the following reasons, runs with these gas mixtures were not made:

(1) Coal gas typically contains less than 1.0 mole percent H_2S and less than 0.05 mole percent COS. The results from runs made with these gases at these levels would have been difficult to assess because of analytical limitations.

(2) Since both H_2S and COS are more soluble in methanol than CO_2, essentially all of these gases would be absorbed for any realistic combination of operating variables in the pilot plant. While early integrated runs showed higher than expected levels of H_2S and COS in the gas exiting the absorber, this was related to the amount of these compounds remaining in the recirculated stripped methanol and not the result of limited column capacity.

(3) The syngas runs were made with CO_2 and N_2 mixtures so that the heat effects associated with the absorption of large amounts of CO_2 could be easily monitored. Although both H_2S and COS have higher heats of solution in methanol than CO_2, the low concentrations of these compounds found in coal gas make the magnitude of this effect relatively

small. It is more realistic to follow CO_2 absorption because of its high concentration in most coal gases.

(4) Mixtures of CO_2 and N_2 were much less expensive to use than large volumes of other gas mixtures. Unless very unusual results were encountered during integrated runs, it was decided to use the gases produced in the gasifier as a feed to the AGRS for multicomponent experiments.

Over the course of the work discussed here, operating procedures and sampling and analytical techniques were continually refined. Improvements were also made in the installation and use of the wall temperature sensors. Although many syngas runs were made, only those that met the material balance closure criteria within 10% and were characterized by steady operation for sufficiently long periods of time were used in parameter estimation. The results of these runs are discussed here and presented in Table 6. Detailed mass balances are given by Ferrell et al in Volume III of this series of reports (1982).

AM-24,25,26,27

These runs were the first in which the wall temperature sensors were used and were made before the gas distributor was repositioned to eliminate end effects. Although several runs made before this time resulted in closed material balances, little information could be gained from them. For the operating ranges studied in these runs most of the entering CO_2 was absorbed and no useful liquid temperature information was obtained.

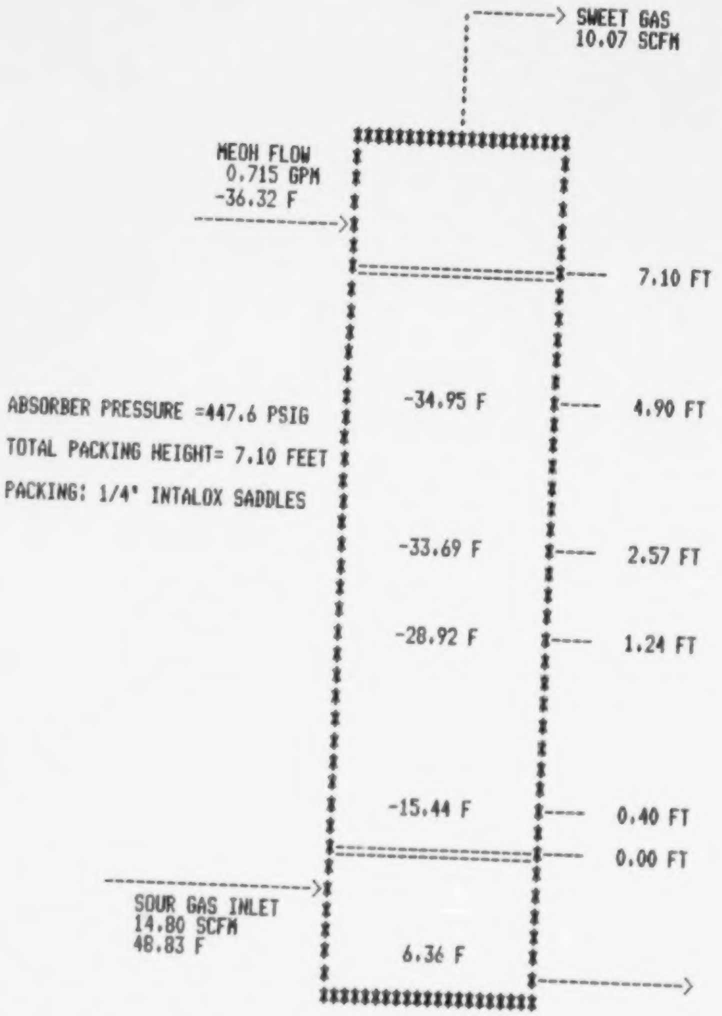
Figures 10 to 13 show all the pertinent information obtained from runs AM-24 to AM-27. Run conditions are summarized in Table 6.

The process parameters that were varied for these runs were the liquid flow rate to the absorber and the column pressure. The results show that over the range of conditions studied, essentially all of the CO_2 fed to the absorber was removed. This indicates that 7.1 feet of packing, in addition to the end effects present at the bottom of the column, were enough to remove the solute to a constant level regardless of liquid flow rate or column pressure. These data reinforced expectations that the column liquid temperature profile was the key to understanding the mass transfer activity in the column.

Figure 10

RUN NUMBER A-M-24
SYNGAS RUN
DATE 3/ 6/1980

COLUMN TEMPERATURE PROFILE


SWEET GAS
10.07 SCFM
MEOH FLOW
0.715 GPM
-36.32 F
7.10 FT
-34.95 F
4.90 FT
-33.69 F
2.57 FT
-28.92 F
1.24 FT
-15.44 F
0.40 FT
0.00 FT
SOUR GAS INLET
14.80 SCFM
48.83 F
6.36 F

ABSORBER PRESSURE =447.6 PSIG

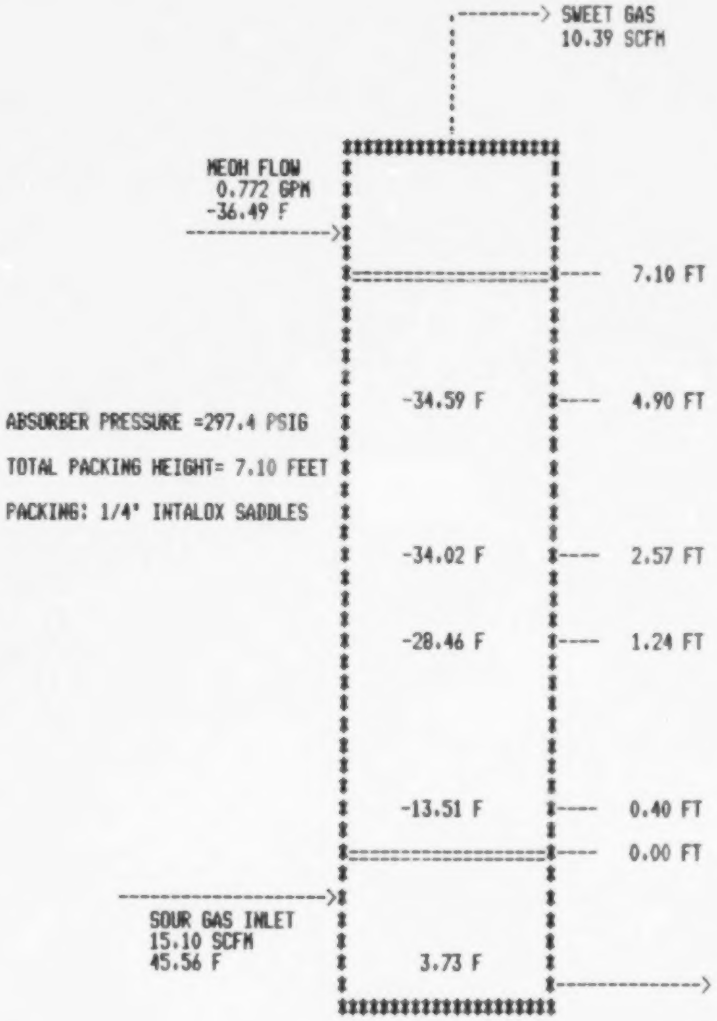
TOTAL PACKING HEIGHT= 7.10 FEET

PACKING: 1/4" INTALOX SADDLES

	SOUR GAS (MOL %)	SWEET GAS (MOL %)
CO2	31.5900	0.0500
H2S	0.0000	0.0000
COS	0.0000	0.0000
MEOH	0.0000	0.9300
H2	0.0000	0.0000
CO	0.0000	0.0000
N2	68.4100	99.0100
CH4	0.0000	0.0000
C2H4	0.0000	0.0000
C2H6	0.0000	0.0000

Figure 11

```
RUN NUMBER A-M-25
  SYNGAS RUN
 DATE    3/26/1980

COLUMN TEMPERATURE PROFILE
------ ----------- -------
```



SWEET GAS
10.39 SCFM
MEOH FLOW
0.772 GPM
-36.49 F
7.10 FT
-34.59 F
4.90 FT
-34.02 F
2.57 FT
-28.46 F
1.24 FT
-13.51 F
0.40 FT
0.00 FT
SOUR GAS INLET
15.10 SCFM
45.56 F
3.73 F

ABSORBER PRESSURE =297.4 PSIG

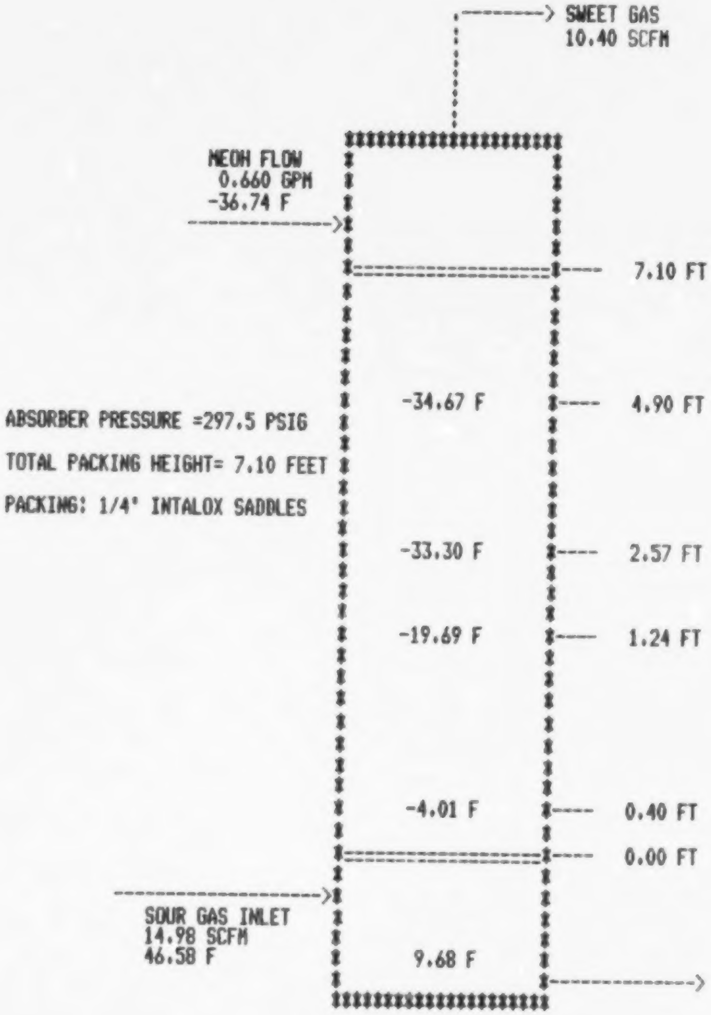
TOTAL PACKING HEIGHT= 7.10 FEET

PACKING: 1/4" INTALOX SADDLES

```
                   SOUR GAS (MOL %)        SWEET GAS (MOL %)
                   ---- ---                ----- ---

CO2                   30.3900                  0.3100
H2S                    0.0000                  0.0000
COS                    0.0000                  0.0000
MEOH                   0.0000                  0.0000
H2                     0.0000                  0.0000
CO                     0.0000                  0.0000
N2                    69.6000                 99.6800
CH4                    0.0000                  0.0000
C2H4                   0.0000                  0.0000
C2H6                   0.0000                  0.0000
```

Figure 12

```
RUN NUMBER A-M-26
 SYNGAS RUN
DATE    4/ 1/1980

COLUMN TEMPERATURE PROFILE
------ ----------- -------
```

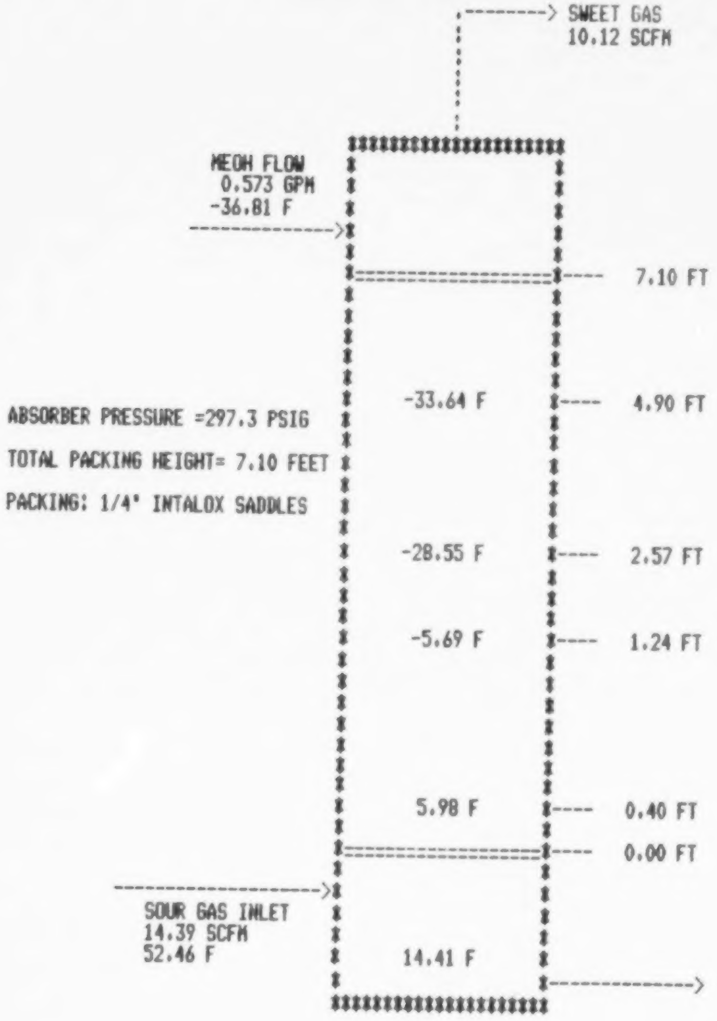


SWEET GAS
10.40 SCFM
MEOH FLOW
0.660 GPM
-36.74 F
7.10 FT
-34.67 F
4.90 FT
ABSORBER PRESSURE =297.5 PSIG
TOTAL PACKING HEIGHT= 7.10 FEET
PACKING: 1/4' INTALOX SADDLES
-33.30 F
2.57 FT
-19.69 F
1.24 FT
-4.01 F
0.40 FT
0.00 FT
SOUR GAS INLET
14.98 SCFM
46.58 F
9.68 F

```
              SOUR GAS (MOL %)         SWEET GAS (MOL %)
              ---- ---                 ----- ---

 CO2              30.4300                  0.1100
 H2S               0.0000                  0.0000
 COS               0.0000                  0.0000
 MEOH              0.0000                  0.0000
 H2                0.0000                  0.0000
 CO                0.0000                  0.0000
 N2               69.5600                 99.8800
 CH4               0.0000                  0.0000
 C2H4              0.0000                  0.0000
 C2H6              0.0000                  0.0000
```

Figure 13

```
RUN NUMBER A-M-27
  SYNGAS RUN
 DATE    4/15/1980

COLUMN TEMPERATURE PROFILE
------ ----------- -------
```


SWEET GAS
10.12 SCFM
MEOH FLOW
0.573 GPM
-36.81 F
7.10 FT
-33.64 F
4.90 FT
ABSORBER PRESSURE =297.3 PSIG
TOTAL PACKING HEIGHT= 7.10 FEET
PACKING: 1/4" INTALOX SADDLES
-28.55 F
2.57 FT
-5.69 F
1.24 FT
5.98 F
0.40 FT
0.00 FT
SOUR GAS INLET
14.39 SCFM
52.46 F
14.41 F

```
              SOUR GAS (MOL %)       SWEET GAS (MOL %)
              ---- ---               ----- ---

CO2              35.5200                0.1300
H2S               0.0000                0.0000
COS               0.0000                0.0000
MEOH              0.0000                0.0000
H2                0.0000                0.0000
CO                0.0000                0.0000
N2               64.4700               99.8600
CH4               0.0000                0.0000
C2H4              0.0000                0.0000
C2H6              0.0000                0.0000
```

TABLE 6

OPERATING CONDITIONS FOR SYNGAS RUNS

RUN	24	25	26	27	32
L (MOLES/HR/FT^2)	69.39	70.43	57.48	50.82	61.18
G (MOLES/FR/FT^2)	17.79	18.16	18.01	17.30	17.31
TG IN (^{o}F)	48.83	45.56	46.58	52.46	57.42
TL IN (^{o}F)	-33.69	-34.02	-33.30	-28.55	-32.94
PRESSURE (PSIG)	450	300	300	300	400
TL OUT (^{o}F)	6.36	3.73	9.68	14.41	11.34
CO$_2$ IN (MOLE %)	31.59	30.39	30.43	35.52	33.73
CO$_2$ OUT (MOLE %)	0.05	0.31	0.11	0.13	0.92
MASS BALANCE	101.8	101.9	103.4	105.0	105.0

RUN	33	34	38	50	55
L (MOLES/HR/FT^2)	72.05	54.38	91.77	127.56	47.91
G (MOLES/HR/FT^2)	16.92	16.99	17.11	17.50	17.83
TG IN (^{o}F)	53.18	61.24	56.48	40.23	57.07
TL IN (^{o}F)	-28.91	-29.33	-18.62	-6.33	-4.17
PRESSURE (PSIG)	400	400	400	250	250
TL OUT (^{o}F)	5.78	15.75	10.70	11.17	29.01
CO$_2$ IN (MOLE %)	34.91	34.54	31.83	32.33	33.13
CO$_2$ OUT (MOLE %)	0.85	1.22	trace	trace	13.06
MASS BALANCE	105.5	104.7	105.0	105.9	93.2

AM-32,33,34

After the gas distributor was repositioned, three runs were made to see if the modification had affected absorber operation. For these runs, the only process parameter that was changed was the liquid flow rate to the absorber. At this point in the study, data showed that the effects of pressure and temperature on CO_2 absorption could be approximated by Henry's law, and that the liquid flow rate (especially its effect on the amount of effective interfacial area) was the key variable.

The results from these runs are summarized in Figures 14, 15, and 16. As before, most of the entering CO_2 was removed even in run AM-34 which used a very low liquid rate. The presence of slightly more CO_2 in the sweet gas may be due to the absence of end effects which had been removed by the column modification discussed previously.

AM-38, AM-50, AM-55

As the study progressed, modeling work showed that the information already obtained from syngas runs could be used in predicting the results from multicomponent runs. Runs AM-38 and AM-50 were made to provide a broader data base with which to determine model parameters. The results are shown in Figures 17, 18, and 19. In addition, a more effective technique for securing the temperature sensors to the column wall had been devised and implemented.

Run AM-55 was made with the intention of having a run at or below the minimum L/G ratio as determined from modeling work. The applicability of the Treybal algorithm (1969) to conditions close to the minimum L/G ratio has been questioned by Stockar and Wilke (1977).

Syngas Summary

These 10 syngas runs cover the range of conditions used in the experimental plan devised for the integrated char and coal runs. The data obtained were used to determine the adjustable parameter created to modify the interfacial area relationships of various researchers. This was necessary because packing sizes as small as the one used in this study are rarely incorporated in mass transfer investigations. The comparison of these runs to model prediction and the technique used to determine the adjustable interfacial parameter are presented in the next section.

Figure 14

```
                         RUN NUMBER A-M-32
                           SYNGAS
                          DATE    6/12/1980

                       COLUMN TEMPERATURE PROFILE
                       ------ ----------- -------

                                 .--------> SWEET GAS
                                 :            9.96 SCFM
                                 :
                                 :
                         ********************
          MEOH FLOW      *                  *
           0.660 GPM     *                  *
          -36.10 F       *                  *
       --------------->  *                  *
                         *==================*----     7.10 FT
                         *                  *
                         *                  *
                         *                  *
                         *    -32.94 F      *----     4.79 FT
ABSORBER PRESSURE =397.2 PSIG               *
TOTAL PACKING HEIGHT= 7.10 FEET             *
PACKING: 1/4" INTALOX SADDLES               *
                         *                  *
                         *    -31.79 F      *----     2.46 FT
                         *                  *
                         *    -23.48 F      *----     1.21 FT
                         *                  *
                         *    -13.93 F      *----     0.79 FT
                         *                  *
                         *     -6.93 F      *----     0.31 FT
                         *==================*----     0.00 FT
       --------------->  *                  *
       SOUR GAS INLET    *                  *
       14.39 SCFM        *                  *
       57.42 F           *     11.34 F      *
                         *                  *-------------->
                         ********************

                          SOUR GAS (MOL %)        SWEET GAS (MOL %)
                          ---- ---                ----- ---

          CO2                 33.7300                 0.9200
          H2S                  0.0000                 0.0000
          COS                  0.0000                 0.0000
          MEOH                 0.0000                 0.0000
          H2                   0.0000                 0.0000
          CO                   0.0000                 0.0000
          N2                  66.2700                99.0800
          CH4                  0.0000                 0.0000
          C2H4                 0.0000                 0.0000
          C2H6                 0.0000                 0.0000
```

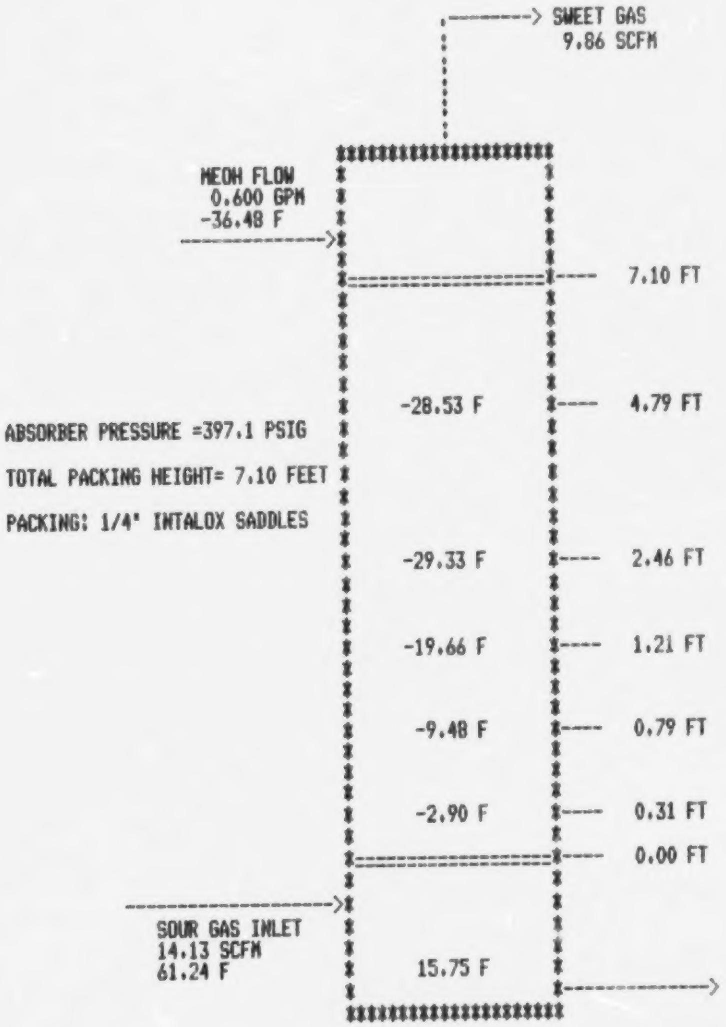
Figure 15

```
                              RUN NUMBER A-M-33
                                SYNGAS RUN
                               DATE    6/19/1980

                           COLUMN TEMPERATURE PROFILE
                           ------ ----------- -------

                                   .--------> SWEET GAS
                                   :            9.76 SCFM
                                   :
                                   :
                                   :
                                   :
                         ********************
              MEOH FLOW  *                  *
               0.780 GPM *                  *
              -35.43 F   *                  *
          -------------->*                  *
                         *                  *
                         *==================*----    7.10 FT
                         *                  *
                         *                  *
                         *                  *
                         *                  *
                         *                  *
                         *    -30.83 F      *----    4.79 FT
ABSORBER PRESSURE =397.2 PSIG               *
                         *                  *
TOTAL PACKING HEIGHT= 7.10 FEET             *
                         *                  *
PACKING: 1/4" INTALOX SADDLES               *
                         *                  *
                         *    -28.91 F      *----    2.46 FT
                         *                  *
                         *                  *
                         *    -27.84 F      *----    1.21 FT
                         *                  *
                         *                  *
                         *    -20.22 F      *----    0.79 FT
                         *                  *
                         *                  *
                         *    -15.09 F      *----    0.31 FT
                         *                  *
                         *==================*----    0.00 FT
      ------------------>*                  *
         SOUR GAS INLET  *                  *
         14.08 SCFM      *                  *
         53.18 F         *     5.78 F       *
                         *                  *-------------->
                         ********************

                           SOUR GAS (MOL %)          SWEET GAS (MOL %)
                           ---- ---                  ----- ---

            CO2               34.5100                   0.8500
            H2S                0.0000                   0.0000
            COS                0.0000                   0.0000
            MEOH               0.0000                   0.0000
            H2                 0.0000                   0.0000
            CO                 0.0000                   0.0000
            N2                65.4900                  99.1500
            CH4                0.0000                   0.0000
            C2H4               0.0000                   0.0000
            C2H6               0.0000                   0.0000
```

RUN NUMBER A-M-34
SYNGAS
DATE 6/19/1980

Figure 16

COLUMN TEMPERATURE PROFILE


SWEET GAS
9.86 SCFM
MEOH FLOW
0.600 GPM
-36.48 F
7.10 FT
-28.53 F
4.79 FT
ABSORBER PRESSURE =397.1 PSIG
TOTAL PACKING HEIGHT= 7.10 FEET
PACKING: 1/4" INTALOX SADDLES
-29.33 F
2.46 FT
-19.66 F
1.21 FT
-9.48 F
0.79 FT
-2.90 F
0.31 FT
0.00 FT
SOUR GAS INLET
14.13 SCFM
61.24 F
15.75 F

	SOUR GAS (MOL %)	SWEET GAS (MOL %)
CO2	34.5400	1.2200
H2S	0.0000	0.0000
COS	0.0000	0.0000
MEOH	0.0000	0.0000
H2	0.0000	0.0000
CO	0.0000	0.0000
N2	65.4500	98.7800
CH4	0.0000	0.0000
C2H4	0.0000	0.0000
C2H6	0.0000	0.0000

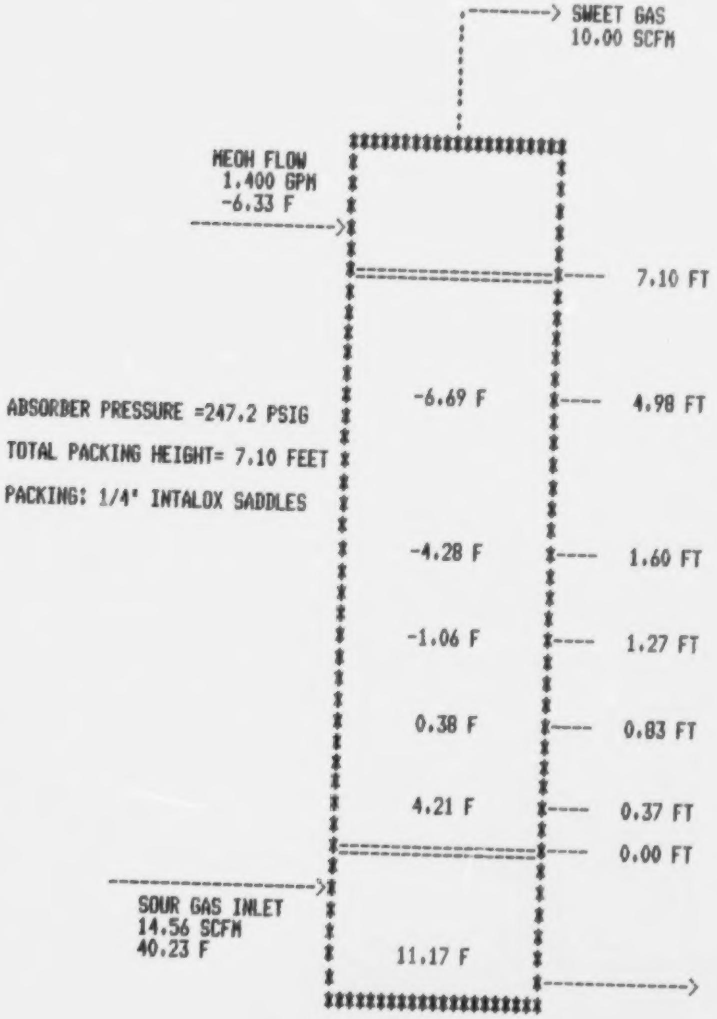
Figure 17

```
                         RUN NUMBER A-M-38
                           SYNGAS
                          DATE    9/10/1980

                       COLUMN TEMPERATURE PROFILE
                       ------ ----------- -------

                                  .--------> SWEET GAS
                                  :            9.76 SCFM
                                  :
                                  :
                       ********************
             MEOH FLOW *                  *
              1.000 GPM*                  *
             -21.13 F  *                  *
       --------------->*                  *
                       *                  *
                       *==================*----     7.10 FT
                       *                  *
                       *                  *
                       *                  *
                       *    -21.13 F      *----     4.79 FT
ABSORBER PRESSURE =397.2 PSIG             *
                       *                  *
TOTAL PACKING HEIGHT= 7.10 FEET           *
                       *                  *
PACKING: 1/4' INTALOX SADDLES             *
                       *                  *
                       *    -18.62 F      *----     2.46 FT
                       *                  *
                       *    -15.75 F      *----     1.21 FT
                       *                  *
                       *    -12.59 F      *----     0.79 FT
                       *                  *
                       *     -5.69 F      *----     0.31 FT
                       *==================*----     0.00 FT
                       *                  *
     ----------------->*                  *
        SOUR GAS INLET *                  *
        14.23 SCFM     *                  *
        56.48 F        *    10.70 F       *
                       *                  *-------------->
                       ********************

                        SOUR GAS (MOL %)        SWEET GAS (MOL %)
                        ---- ---                ----- ---

            CO2             31.9100                  0.0000
            H2S              0.0000                  0.0000
            COS              0.0000                  0.0000
            MEOH             0.0000                  0.0000
            H2               0.0000                  0.0000
            CO               0.0000                  0.0000
            N2              68.0900                100.0000
            CH4              0.0000                  0.0000
            C2H4             0.0000                  0.0000
            C2H6             0.0000                  0.0000
```

Figure 18

RUN NUMBER A-M-50
SYNGAS RUN
DATE 2/12/1981

COLUMN TEMPERATURE PROFILE

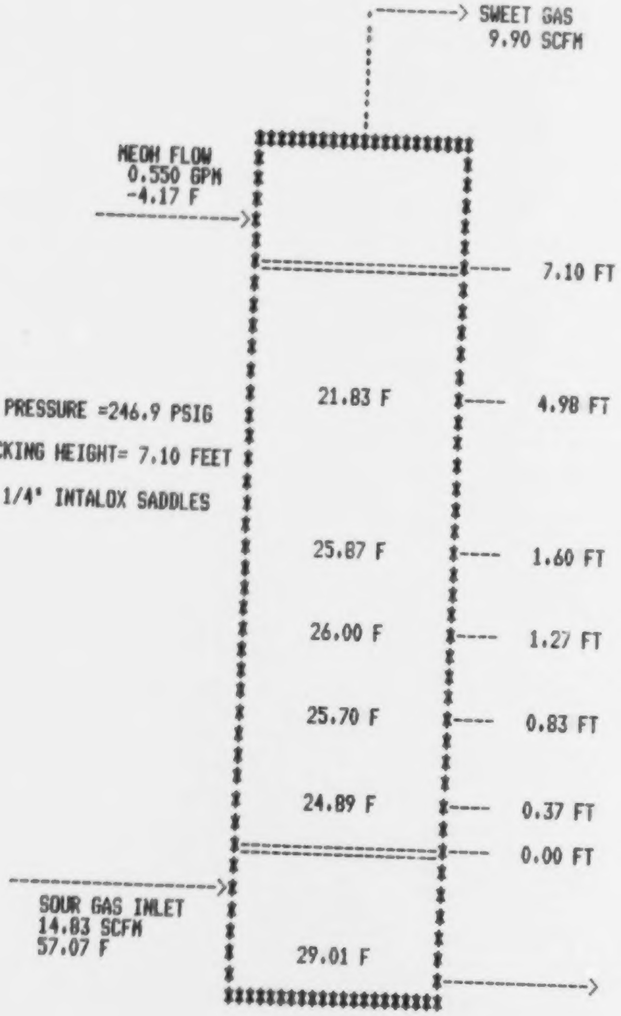

SWEET GAS
10.00 SCFM
MEOH FLOW
1.400 GPM
-6.33 F
7.10 FT
-6.69 F
4.98 FT
ABSORBER PRESSURE =247.2 PSIG
TOTAL PACKING HEIGHT= 7.10 FEET
PACKING: 1/4" INTALOX SADDLES
-4.28 F
1.60 FT
-1.06 F
1.27 FT
0.38 F
0.83 FT
4.21 F
0.37 FT
0.00 FT
SOUR GAS INLET
14.56 SCFM
40.23 F
11.17 F

	SOUR GAS (MOL %)	SWEET GAS (MOL %)
CO2	32.2300	0.0000
H2S	0.0000	0.0000
COS	0.0000	0.0000
MEOH	0.0000	0.0000
H2	0.0000	0.0000
CO	0.0000	0.0000
N2	67.7600	100.0000
CH4	0.0000	0.0000
C2H4	0.0000	0.0000
C2H6	0.0000	0.0000

Figure 19

RUN NUMBER A-M-55
SYNGAS
DATE 4/14/1981

COLUMN TEMPERATURE PROFILE


SWEET GAS
9.90 SCFM
MEOH FLOW
0.550 GPM
-4.17 F
7.10 FT
21.83 F
4.98 FT
25.87 F
1.60 FT
26.00 F
1.27 FT
25.70 F
0.83 FT
24.89 F
0.37 FT
0.00 FT
SOUR GAS INLET
14.83 SCFM
57.07 F
29.01 F

ABSORBER PRESSURE =246.9 PSIG

TOTAL PACKING HEIGHT= 7.10 FEET

PACKING: 1/4" INTALOX SADDLES

	SOUR GAS (MOL %)	SWEET GAS (MOL %)
CO2	33.1300	13.0600
H2S	0.0000	0.0000
COS	0.0000	0.0000
MEOH	0.0000	0.0000
H2	0.0000	0.0000
CO	0.0000	0.0000
N2	66.8600	0.0000
CH4	0.0000	86.9300
C2H4	0.0000	0.0000
C2H6	0.0000	0.0000

COMPARISON OF EXPERIMENTAL DATA TO SIMPAK

The 10 syngas runs were used to determine the accuracy of the mass transfer coefficient correlations and the interfacial area relationships used in the model. Because four options were available for evaluating these quantities, it was possible that the results obtained for each set of transfer coefficient and interfacial area correlations would be different.

The method used to determine the accuracy of these correlations involved the measured liquid temperature profiles from the syngas runs. The procedure was as follows:

1. After a SYNGAS run was completed, the data were reduced and the mass balance tested. The CO_2 removal efficiency was calculated and the liquid temperature profile recorded.
2. The computer model SIMPAK or MCOMP was executed using the experimental process conditions. The CO_2 removal efficiency was entered into the program. With this information, the liquid temperature profile was calculated as a function of the packed height.
3. The mass transfer coefficient-interfacial area options were evaluated to see which resulted in the best fit of the computed liquid temperature profiles to the experimental data.

In applying the above procedure to the 10 syngas runs, the model failed to converge for AM-27 and AM-55. Further analysis of these runs is discussed later in the section on minimum L/G ratios.

Of the three sets of transfer coefficient and interfacial area relations considered, only the correlation of Onda et al (1968) had been developed using data for packing sizes smaller than 1/2 inch. For this reason, it was chosen as the correlation to be used in the initial model comparisons.

Comparisons ot experimental data with model predictions were made with the following options chosen in the input to SIMPAK:

Binary gas diffusivity option (Wilke-Lee Modification to the Chapman-Enskog)	2
Binary liquid diffusivity option (Wilke-Chang Modification to the Stokes-Einstein equation)	1
Transfer coefficient option (Correlation of Onda et al.)	2
Packing increments	50

One packing increment is defined as the total height of packing required to remove a specified amount of CO_2 or any other related key component. For 50 packing increments, each increment is equivalent to the height required to remove 1/50 of the specified amount of the key component and is affected by the conditions in the packing. The heights calculated for all increments are summed to yield the total height of packing. Fifty increments were chosen as a compromise between computer time and accuracy; the larger the number of increments the more closely the gradients calculated approach a true differential.

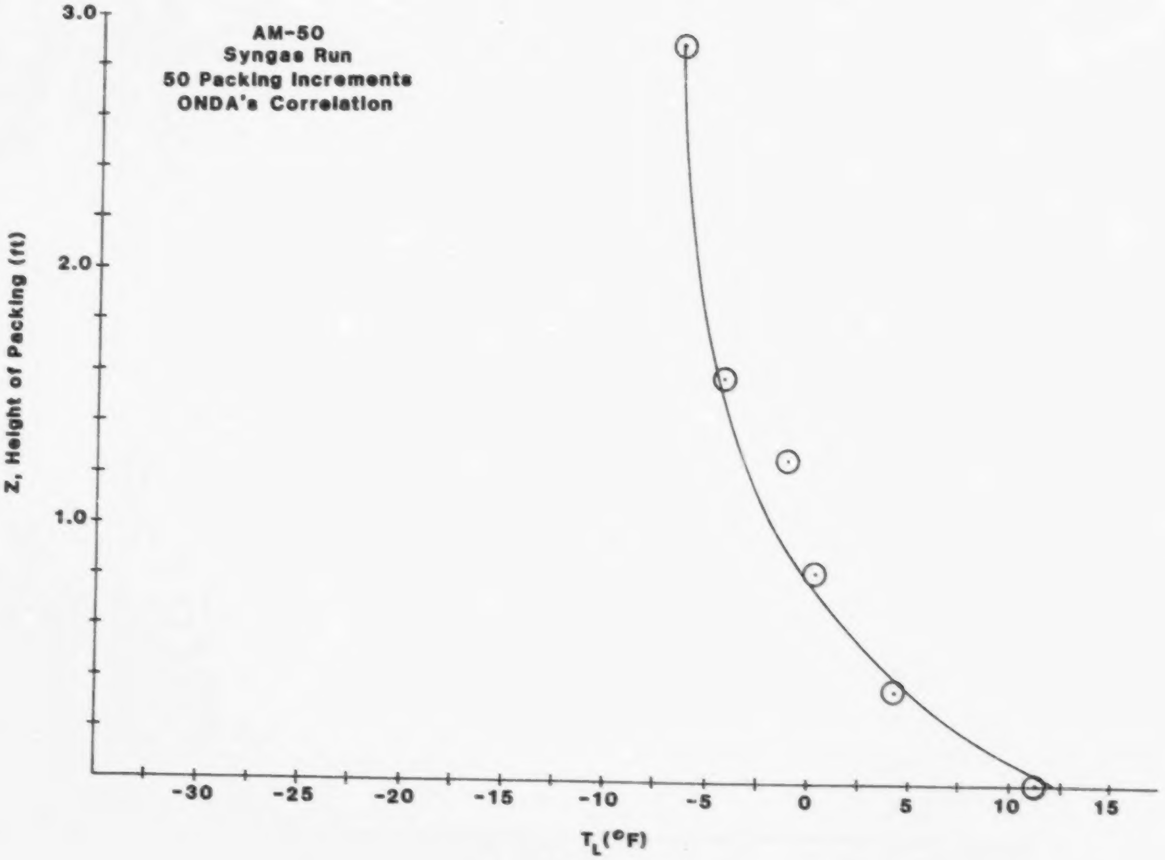
It should be noted that the inlet liquid temperature used in the model input was not always the temperature of the liquid entering the column at the top of the packing. Because of the distance of this temperature sensor from the actual column inlet, some deviation was usually noted between the temperature at the five foot level of packing and the inlet temperature. This was especially true at low liquid flow rates. For this reason, the inlet temperature to the absorber was taken to be the reading at the five foot level in the preliminary model run. If the computed height was much less than five feet, the value of the temperature sensor at that computed height was used in the next model run. Although a more positive determination of the inlet liquid temperature is desirable, the method used here gave a more realistic picture of column performance than would have been the case if the nominal inlet liquid temperatures had been used.

In general, model predictions compared well with experimental data. The discussion of the results of these runs begins with AM-50 since it took place after many of the problems with the wall temperature sensors had been solved.

AM-50

The experimental results from AM-50 can be found in Table 7 and in Figure 20, which show excellent agreement between the computed liquid temperature profile and the experimental data. The fact that the run was

Figure 20


AM-50
Syngas Run
50 Packing Increments
ONDA's Correlation
Z, Height of Packing (ft)
3.0
2.0
1.0
-30
-25
-20
-15
-10
-5
0
5
10
15
T_L(°F)

made with a high liquid flow rate, 127 lb-moles/hr/ft^2, probably promoted good heat transfer that may have improved the accuracy of the wall temperature sensors.

AM-38

Because excellent agreement was achieved by using the Onda correlation it was also used for the other runs. The results from AM-38 are shown in Figure 21. Again, excellent agreement between model and experimental data was observed. The inlet liquid temperature was assumed to be equal to the value obtained from the sensor at 2.46 feet of packing. This was done because in the preliminary model run, this was approximately the required height to achieve the specified recovery of CO_2.

AM-32,33,34

The liquid temperature profiles from these runs are compared to computed profiles in Figures 22, 23, and 24. Although there were difficulties in securing the sensors to the column wall in the runs, very good agreement is noted for runs AM-32 and AM-34. The lowest temperature sensor in each of these runs was found to be improperly attached which probably explains its deviation from the predicted values. In AM-33, both the lowest sensor and the sensor located at 1.2 feet appeared not to have been properly fastened. Fairly good agreement is still evident.

AM-24,25,26

These three runs were made before the gas distributor was moved to eliminate end effects at the bottom of the column. For these three runs, comparisons of computed to measured liquid temperature profiles reflect the influence of the end effects as seen in Figures 25 to 27. Wall temperatures are all colder than the predicted profiles. The conclusion that may be drawn is that some CO_2 was absorbed below the bottom of the packing. This would cause the thermal effect from the heat of solution of CO_2 in methanol to be reduced and the predicted temperature profile, which assumes that all the CO_2 is absorbed in the packing, would show higher temperatures than those measured during the syngas run.

Discussion

Agreement between measured syngas temperature profiles and calculated values showed that the model satisfactorily predicted column performance. The correlations suggested by Onda et al (1968) for mass transfer

Figure 21

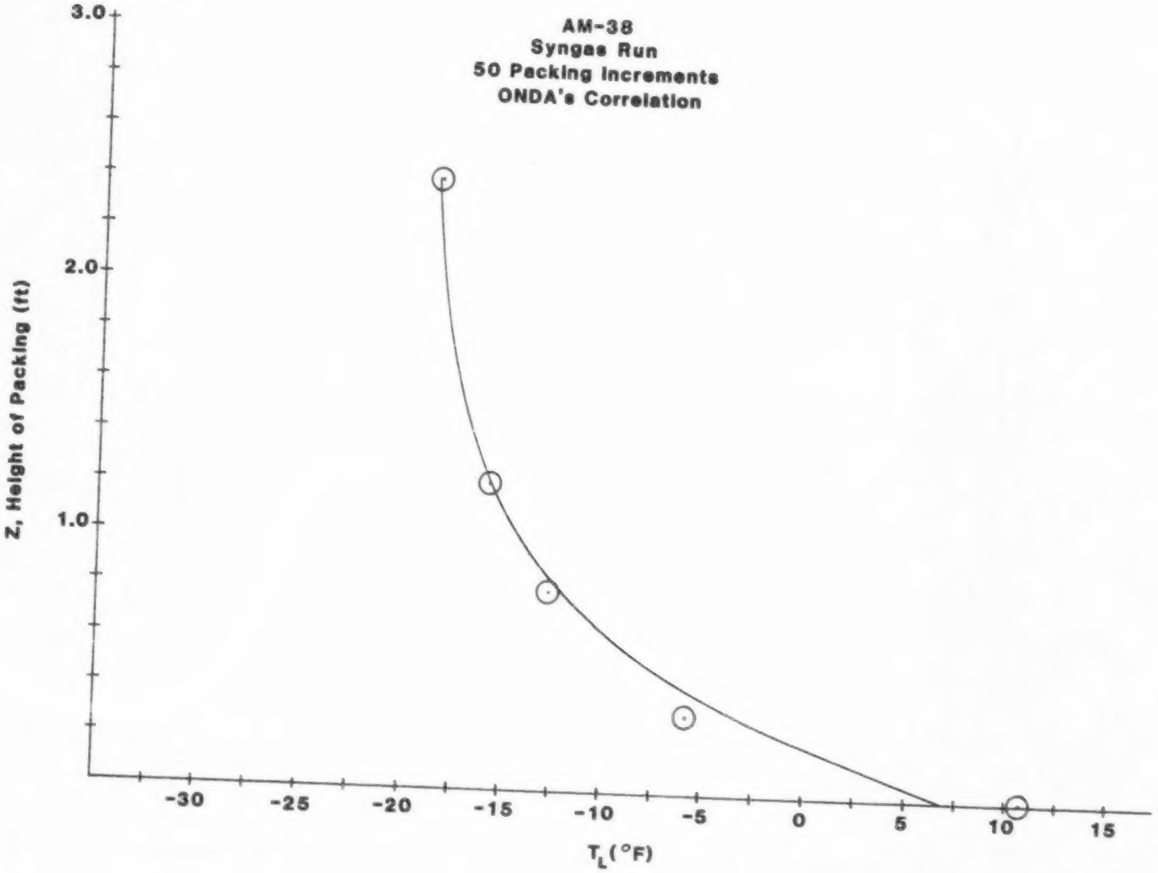
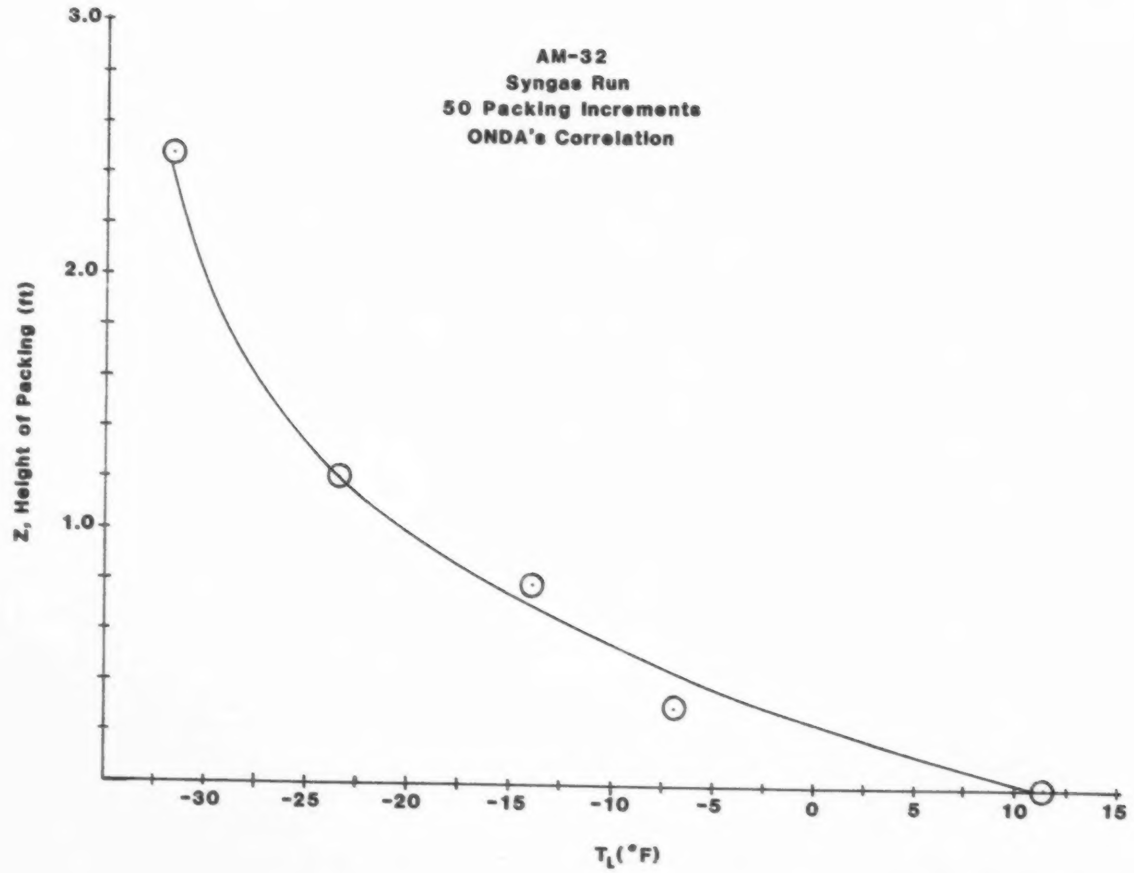

AM-38
Syngas Run
50 Packing Increments
ONDA's Correlation
Z, Height of Packing (ft)
3.0
2.0
1.0
-30
-25
-20
-15
-10
-5
0
5
10
15
T_L (°F)

Figure 22



AM-32
Syngas Run
50 Packing Increments
ONDA's Correlation

Figure 23

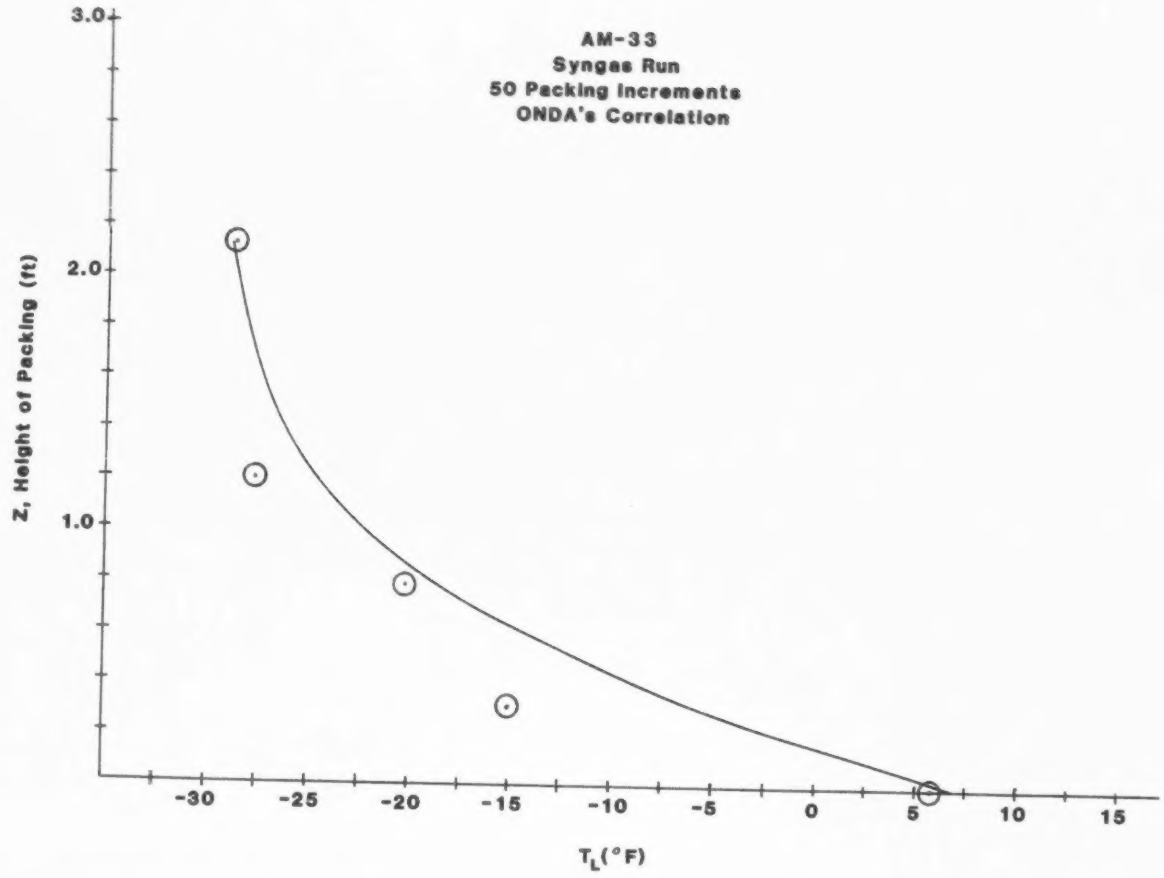

AM-33
Syngas Run
50 Packing Increments
ONDA's Correlation
Z, Height of Packing (ft)
3.0
2.0
1.0
-30
-25
-20
-15
-10
-5
0
5
10
15
T_L(°F)

Figure 24

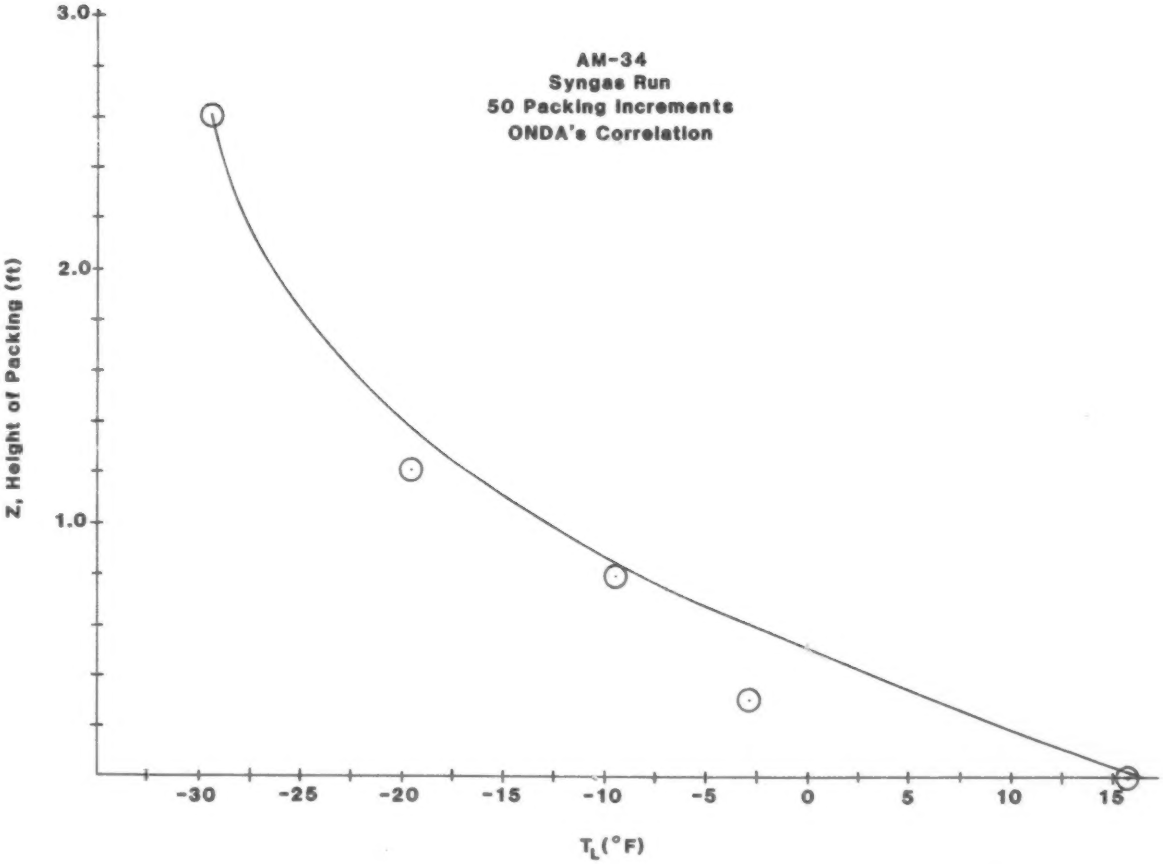

AM-34
Syngas Run
50 Packing Increments
ONDA's Correlation
Z, Height of Packing (ft)
3.0
2.0
1.0
-30
-25
-20
-15
-10
-5
0
5
10
15
T_L(°F)

Figure 25

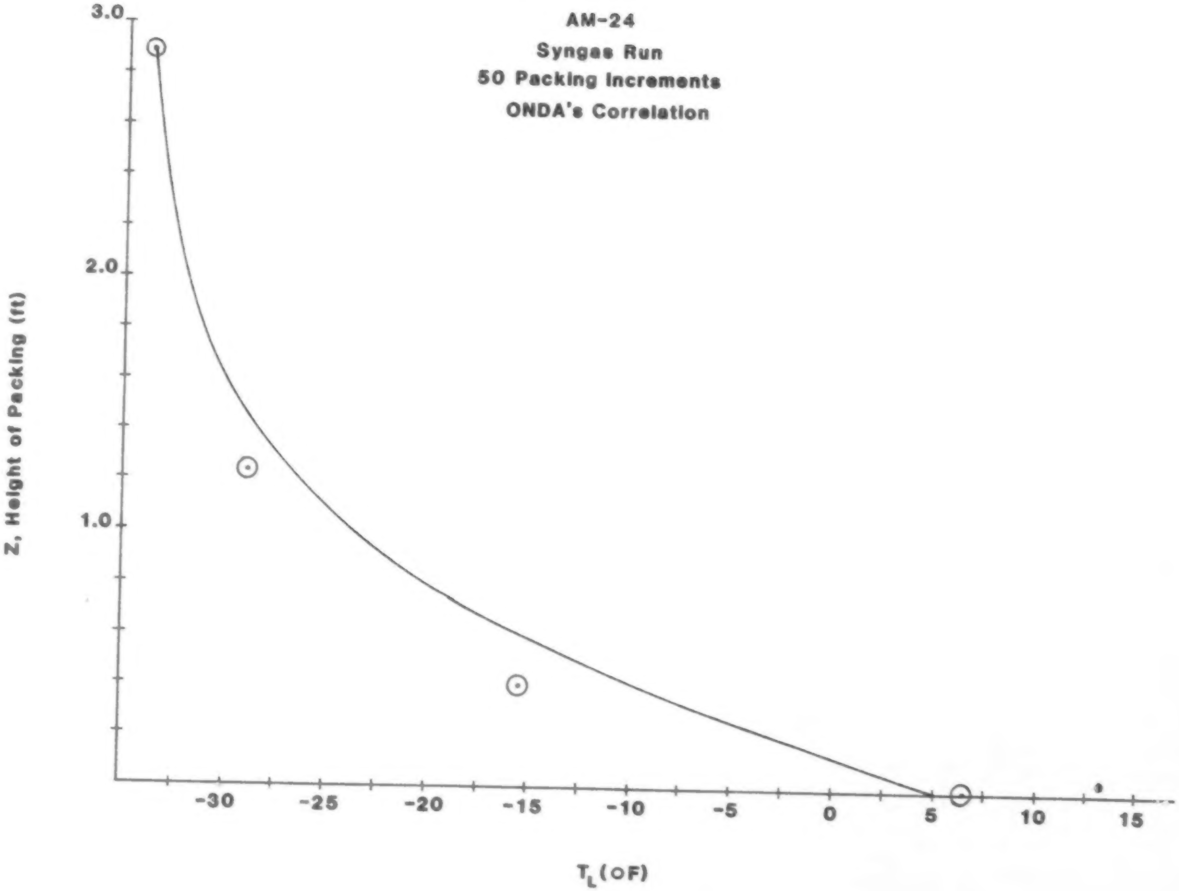

AM-24
Syngas Run
50 Packing Increments
ONDA's Correlation
Z, Height of Packing (ft)
3.0
2.0
1.0
-30
-25
-20
-15
-10
-5
0
5
10
15
T_L (oF)

Figure 26

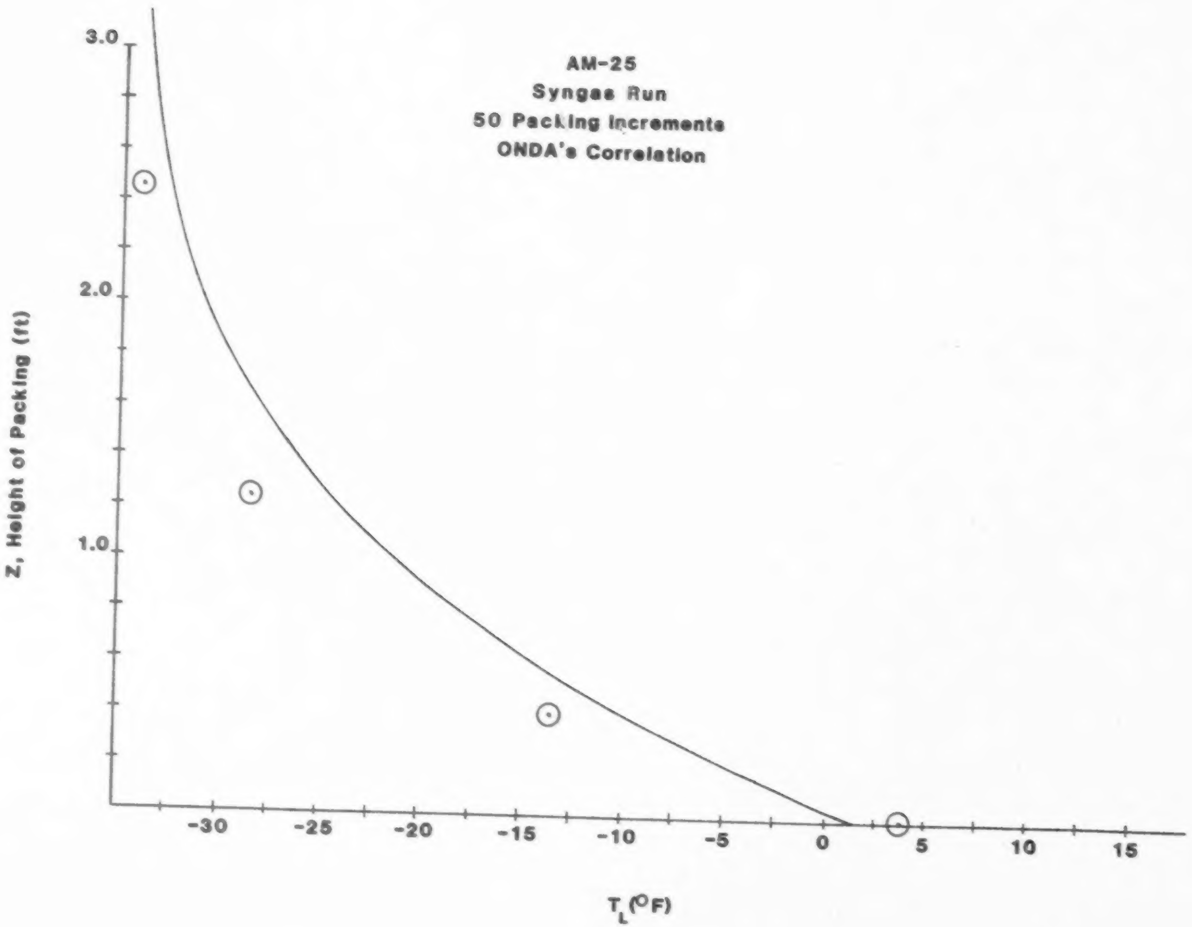
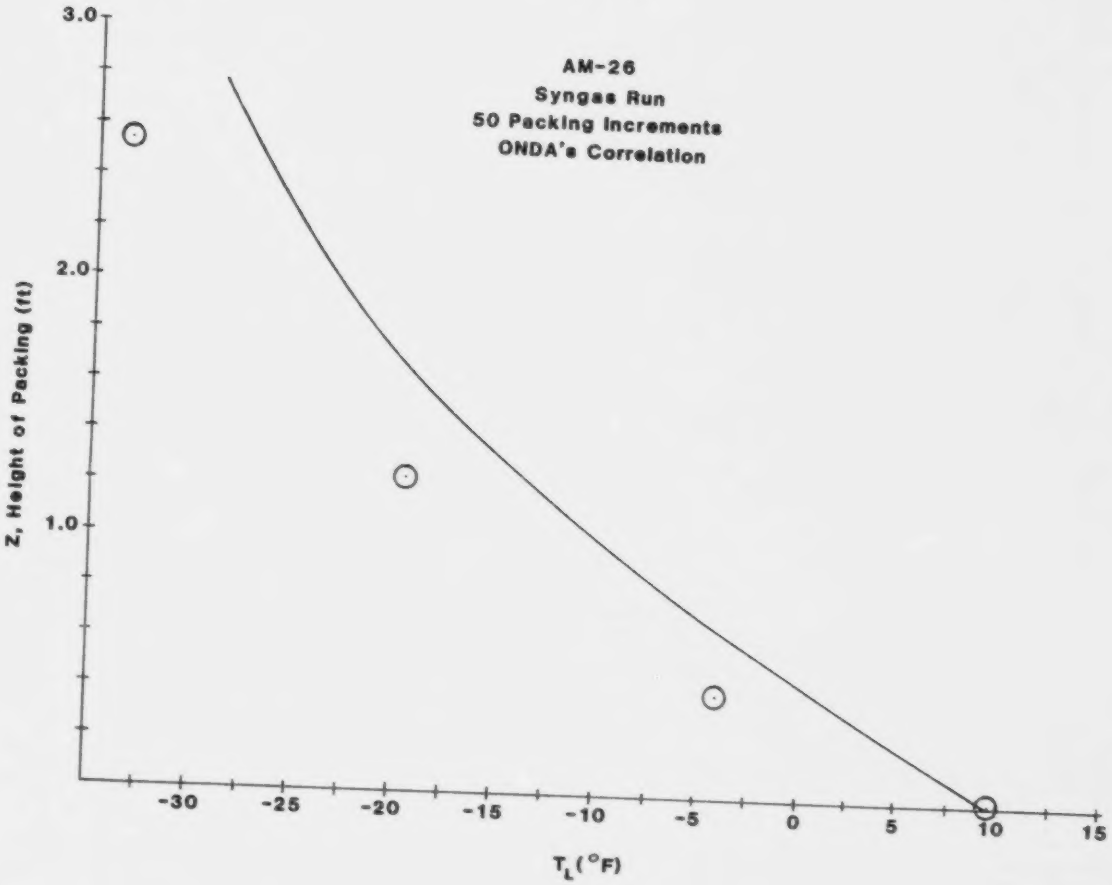

AM-25
Syngas Run
50 Packing Increments
ONDA's Correlation
Z, Height of Packing (ft)
3.0
2.0
1.0
-30
-25
-20
-15
-10
-5
0
5
10
15
T_L(°F)

Figure 27


AM-26
Syngas Run
50 Packing Increments
ONDA's Correlation

coefficients and interfacial areas accurately described column performance. Examination of the plots of the height of packing versus the liquid temperature show that the shape of the predicted curve closely follows the experimental data. Differences between model predictions and experimental measurements were substantially reduced in later runs, especially those that were integrated. This would seem to indicate that the differences between model and experiment noted for these early runs were due to the unrefined experimental technique.

Table 7 summarizes syngas run results and SIMPAK predictions of the height of packing required to accomplish the specified recovery of the entering solute. The inlet liquid temperature given is that used in the model input. In all cases where the model converged, much less than the 7.1 feet of packing was required to achieve the specified CO_2 recovery. Table 8 compares predicted and actual liquid temperatures. The fact that these compare closely confirms the accuracy of the flow meter calibrations, the sampling and analytical techniques and the validity of the assumptions made in estimating the heat of solution of CO_2 in methanol from Henry's law constants. The question that still remains is whether these results can be extended and used to predict multicomponent system behavior. The use of the program MCOMP for this purpose is presented in the next section.

TABLE 7

SYNGAS RUN RESULTS

RUN	L	T_L	G	T_G	P	CO_2 RECOVERY	COMPUTED HEIGHT OF PACKING (ONDA)
24	69.39	-33.69	17.79	48.83	450	0.999	2.874
25	70.43	-34.02	18.16	45.56	300	0.993	3.331
26	57.48	-33.30	18.01	46.58	300	0.997	4.544
27	50.82	-28.55	17.30	52.46	300	0.998	NO CONVERGENCE
32	61.18	-31.79	17.31	57.42	400	0.981	2.447
33	72.05	-28.91	16.92	53.18	400	0.983	2.135
34	54.38	-29.33	16.99	61.24	400	0.975	2.613
38	91.77	-18.62	17.11	56.48	400	0.998	2.403
50	127.56	- 6.33	17.50	40.23	250	0.998	2.898
55	47.91	- 4.17	17.83	57.07	250	0.701	NO CONVERGENCE

L	LB-MOLES/HR/FT^2
G	LB-MOLES/HR/FT^2
T_L	DEGREES F
T_G	DEGREES F
P	PSIG

HEIGHT OF PACKING IN FEET

TABLE 8

COMPARISON OF PREDICTED AND EXPERIMENTAL OUTLET LIQUID TEMPERATURES

RUN	T_L IN	T_L OUT	COMPUTED T_L OUT
24	-33.69	6.36	6.91
25	-34.02	3.73	1.53
26	-33.30	9.68	9.33
27	-28.55	14.41	----
32	-31.79	11.34	9.34
33	-28.91	5.78	6.78
34	-29.33	15.75	17.11
38	-18.62	10.70	7.69
50	- 6.33	11.17	12.03
55	- 4.17	29.01	32.18

WHERE ALL TEMPERATURES ARE DEGREES F

COMPARISON OF EXPERIMENTAL DATA TO MCOMP

Using the method established for comparing results from SYNGAS runs to the prediction from the program SIMPAK, results from integrated runs were compared to predictions from the computer program MCOMP. For each integrated run, the computed liquid temperature profile was compared to the experimental data. The measured outlet gas compositions were also compared to predicted results. Results are presented in Tables 9 and 10. Table 10 shows a comparison of the predicted and actual outlet liquid temperatures. Agreement of the two values is an indication of the accuracy of the adiabatic assumption as well as a check on sampling and analytical techniques.

In all cases, the correlation of Onda et al (1968) was used to estimate the mass transfer coefficient and interfacial area. Options used in the model calculations were chosen in the model input as follows:

Binary gas diffusivity option (Wilke-Lee Modification to the Chapman-Enskog equation)	2
Binary liquid diffusivity option (Wilke-Chang Modification to the Stokes-Einstein equation)	1
Transfer Coefficient option (Correlation of Onda et al.)	2
Liquid Mixture Diffusivity option (Modification of Holmes et al. to the Wilke-Chang equation)	2
Number of Packing Increments	10.0

Because MCOMP handles up to 13 components, considerably more computation time was required than was the case with SIMPAK. For this reason, 10 packing increments were chosen to economize on computer time. The effect of the number of packing increments on model prediction is discussed later.

In all the comparisons of model predictions to experimental results, CO_2 was chosen as the key component. While any of the components factored into the model could be used as the key component, the absorption of CO_2 could be followed most easily through the column liquid temperature profile. The other components were either sparingly soluble in methanol or present in such small quantities that their absorption did not affect the measured temperature profile.

Gases From Char

Integrated runs AMI-30, 35, 36, and 37 were made with a feed gas generated from the gasification of a Western Kentucky coal char. The results from these runs are discussed by Kelly et al (1980). Gases produced from char gasification typically contain no hydrocarbons with molecular weights higher than methane. This particular char contained fairly high levels of sulfur so that relatively large amounts of H_2S and COS were produced. The results from these runs are presented in Figures 28 to 35.

In all cases, the model prediction of MCOMP compares well with the actual liquid temperature profile and outlet gas composition. In these runs, the transfer of eight compounds, including the solvent, was considered. The difference between the actual and predicted H_2S and COS outlet gas concentrations was attributed to the presence of both of these compounds in the recirculated methanol fed to the absorber. This resulted from the inefficient stripping of these compounds from the rich methanol and led to a reduced driving force at the top of the absorber.

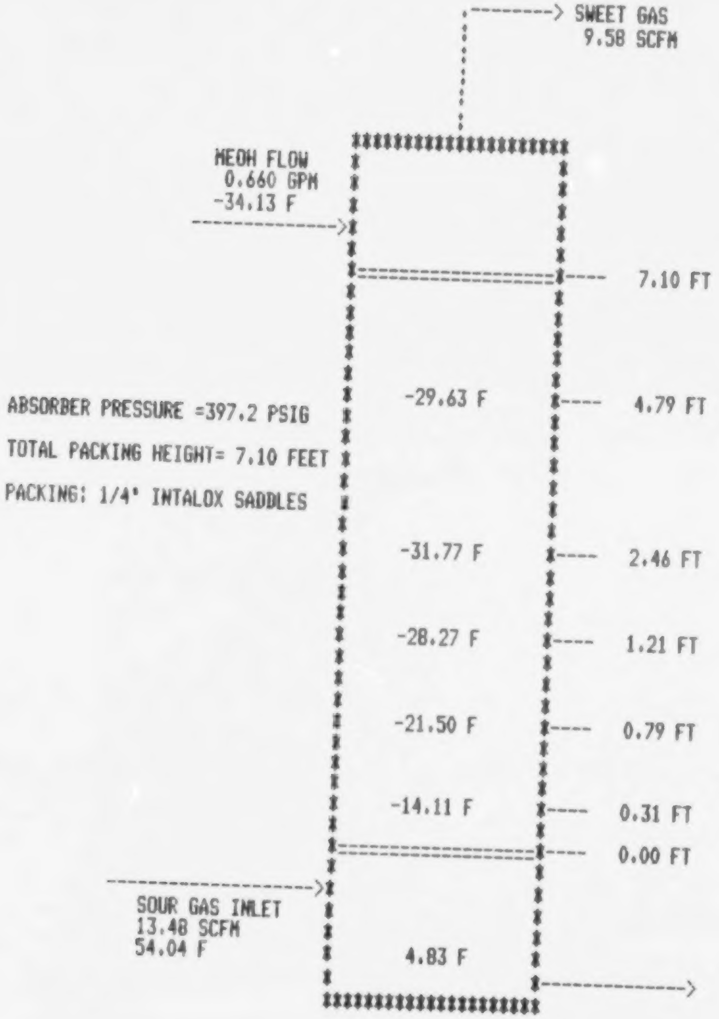
Gases From Coal

Integrated runs AMI-43, 44, 45, 47, 52, 53, 57, 59, and 60 were made using a feed gas generated by the gasification of a New Mexico sub-bituminous coal. The experimental results from these runs are discussed by Rousseau et al (1981). Gases produced from gasification of this particular coal were characterized by fairly low amounts of H_2S and COS, and relatively high levels of both aromatic and aliphatic hydrocarbons. Only those compounds that could be analyzed with some accuracy and that were important in mass balance calculations were included in the computer model runs. Comparison of experimental data to predicted results for these runs can be found in Figures 36 to 53.

In general, the computer prediction compared well with the experimental data. Differences between actual and predicted H_2S and COS concentrations can be explained by the factors cited previously. The difference between the computed and measured outlet gas compositions of ethane and ethylene may be attributed to deviations from Henry's law which had been assumed to hold in the calculation. Although the presence of propane, propylene, and butane was noted in the product gas from the gasifier, analyses for these compounds were not performed routinely and, therefore, not used in the program input. Their total concentration amounted to approximately 0.2 mole percent of the product gas. Results from all integrated runs are listed in Table 9 and 10.

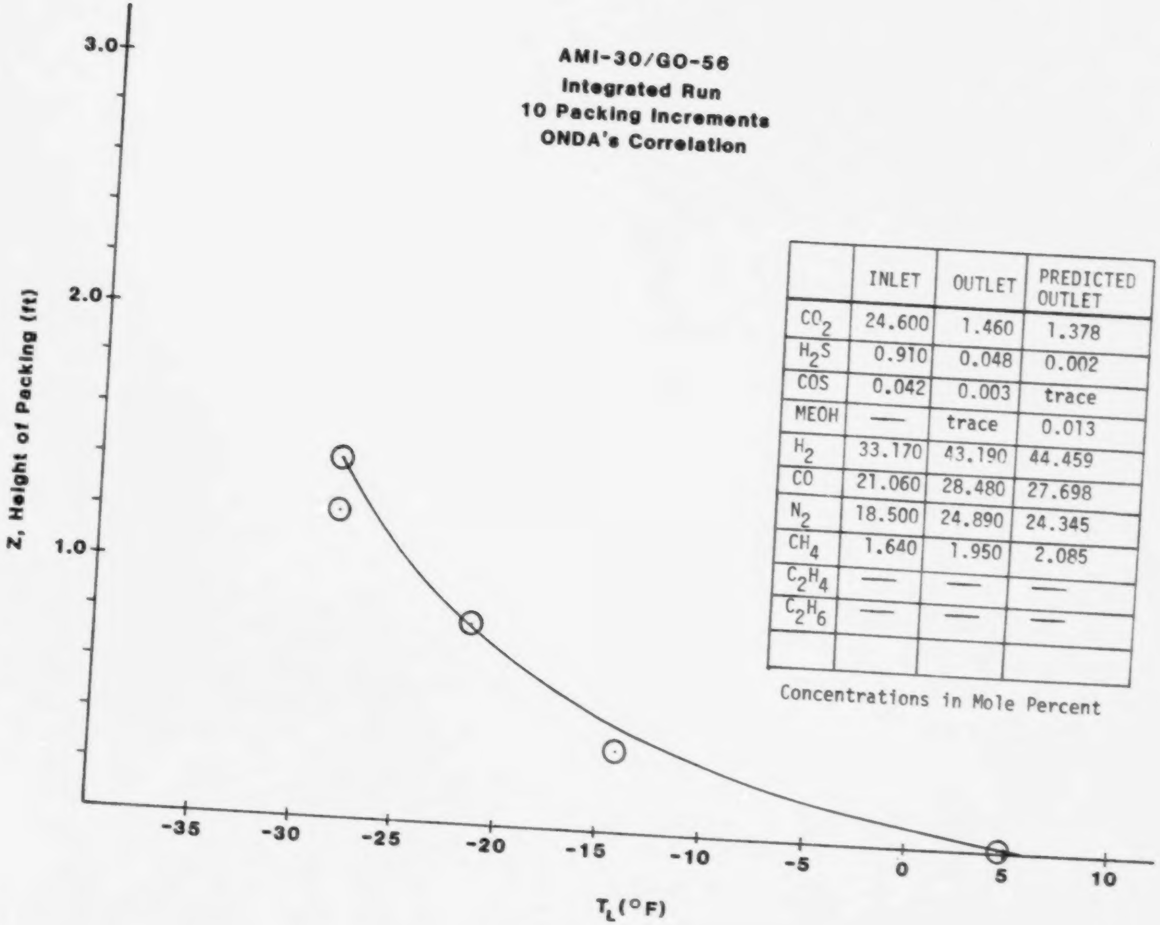
Figure 28

```
RUN NUMBER A-M-30
  INTEGRATED
DATE     5/28/1980

COLUMN TEMPERATURE PROFILE
------ ----------- -------
```


SWEET GAS
9.58 SCFM
MEOH FLOW
0.660 GPM
-34.13 F
7.10 FT
-29.63 F
4.79 FT
ABSORBER PRESSURE =397.2 PSIG
TOTAL PACKING HEIGHT= 7.10 FEET
PACKING: 1/4" INTALOX SADDLES
-31.77 F
2.46 FT
-28.27 F
1.21 FT
-21.50 F
0.79 FT
-14.11 F
0.31 FT
0.00 FT
SOUR GAS INLET
13.48 SCFM
54.04 F
4.83 F

```
              SOUR GAS (MOL %)        SWEET GAS (MOL %)
              ---- ---                ----- ---

CO2            24.6000                 1.4600
H2S             0.9096                 0.0476
COS             0.0423                 0.0032
MEOH            0.0000                 0.0000
H2             33.1700                43.1900
CO             21.0600                28.4800
N2             18.5000                24.8900
CH4             1.6400                 1.9500
C2H4            0.0000                 0.0000
C2H6            0.0000                 0.0000
```

Figure 29



AMI-30/GO-56
Integrated Run
10 Packing Increments
ONDA's Correlation

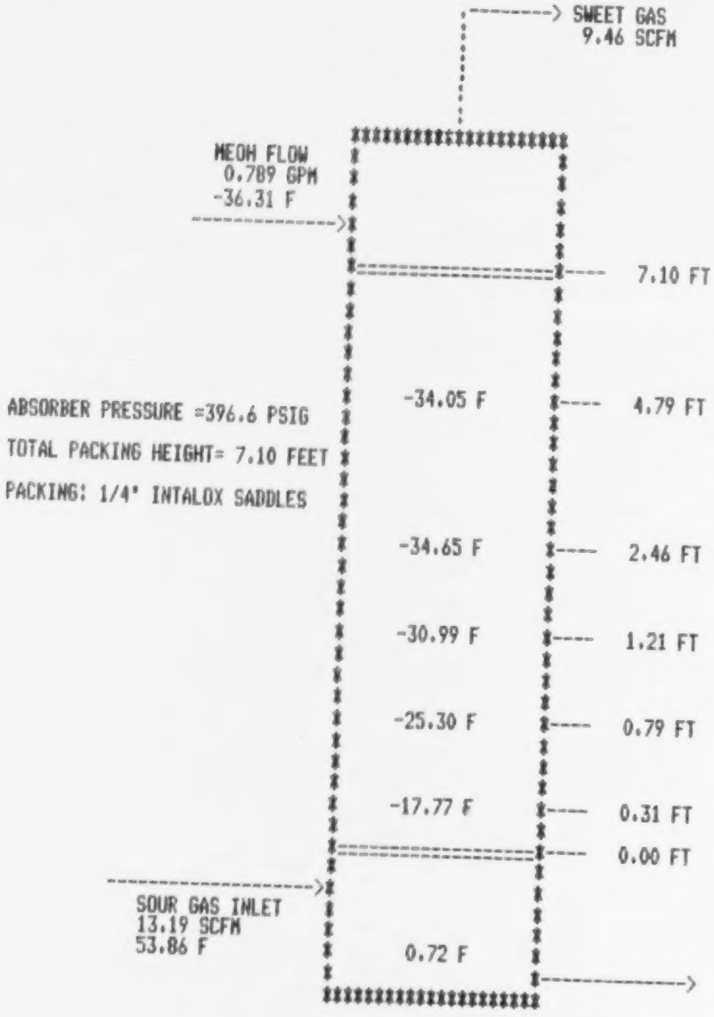
	INLET	OUTLET	PREDICTED OUTLET
CO_2	24.600	1.460	1.378
H_2S	0.910	0.048	0.002
COS	0.042	0.003	trace
MEOH	—	trace	0.013
H_2	33.170	43.190	44.459
CO	21.060	28.480	27.698
N_2	18.500	24.890	24.345
CH_4	1.640	1.950	2.085
C_2H_4	—	—	—
C_2H_6	—	—	—

Concentrations in Mole Percent

Figure 30

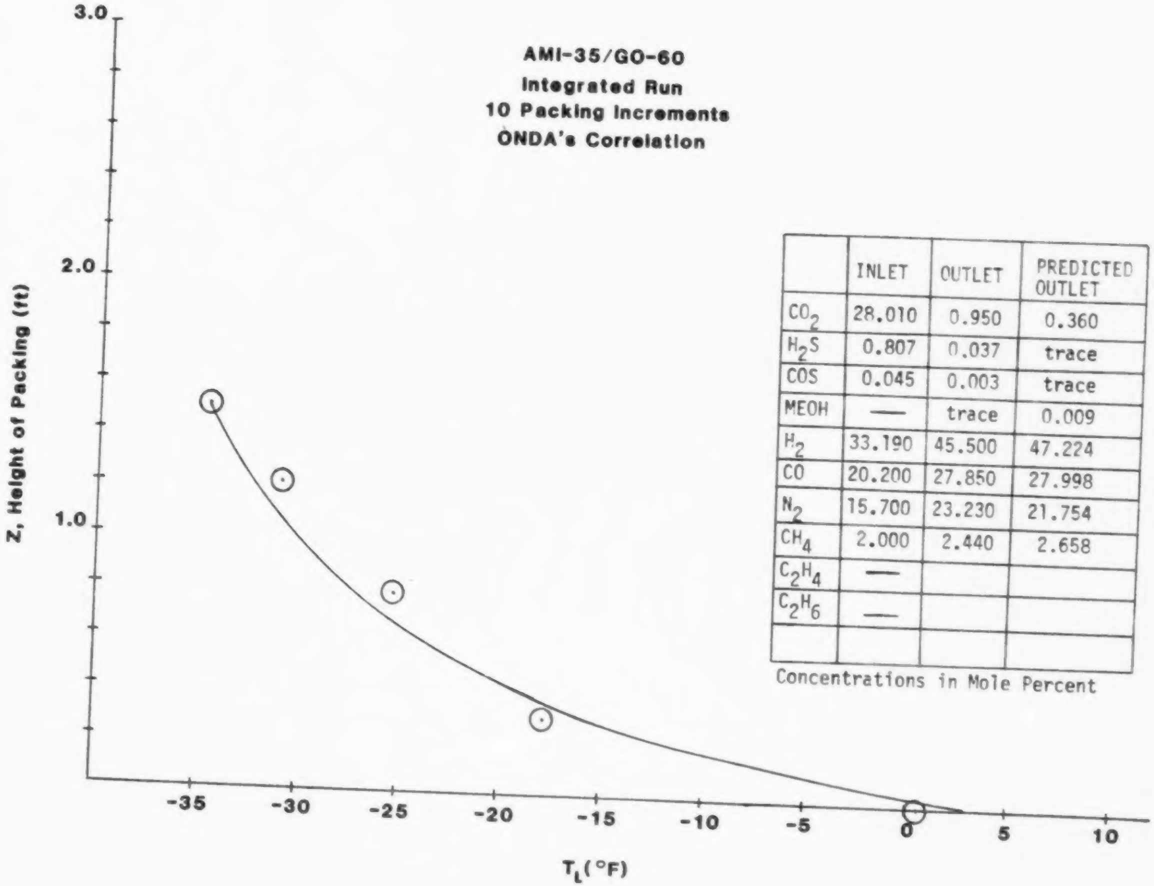
RUN NUMBER A-M-35
INTEGRATED RUN
DATE 6/26/1980

COLUMN TEMPERATURE PROFILE


SWEET GAS
9.46 SCFM
MEOH FLOW
0.789 GPM
-36.31 F
7.10 FT
-34.05 F
4.79 FT
ABSORBER PRESSURE =396.6 PSIG
TOTAL PACKING HEIGHT= 7.10 FEET
PACKING: 1/4" INTALOX SADDLES
-34.65 F
2.46 FT
-30.99 F
1.21 FT
-25.30 F
0.79 FT
-17.77 F
0.31 FT
0.00 FT
SOUR GAS INLET
13.19 SCFM
53.86 F
0.72 F

	SOUR GAS (MOL %)	SWEET GAS (MOL %)
CO2	28.0100	0.9500
H2S	0.8072	0.0371
COS	0.0450	0.0034
MEOH	0.0000	0.0000
H2	33.1900	45.5000
CO	20.2000	27.8500
N2	15.7000	23.2300
CH4	2.0100	2.4400
C2H4	0.0000	0.0000
C2H6	0.0000	0.0000

Figure 31



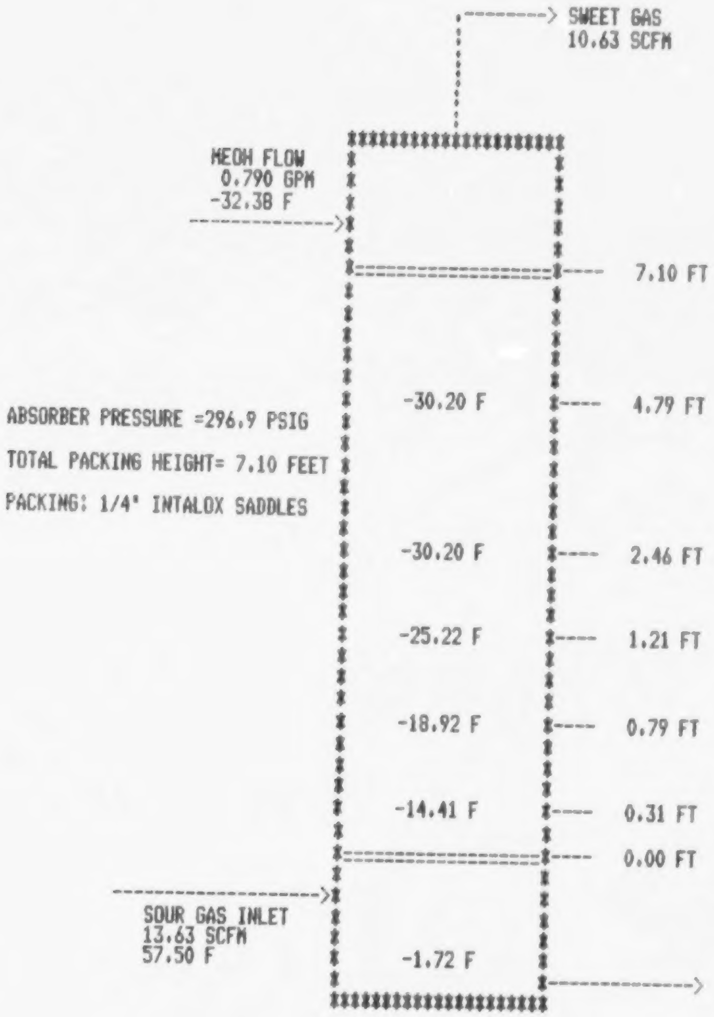
	INLET	OUTLET	PREDICTED OUTLET
CO_2	28.010	0.950	0.360
H_2S	0.807	0.037	trace
COS	0.045	0.003	trace
MEOH	—	trace	0.009
H_2	33.190	45.500	47.224
CO	20.200	27.850	27.998
N_2	15.700	23.230	21.754
CH_4	2.000	2.440	2.658
C_2H_4	—		
C_2H_6	—		

Concentrations in Mole Percent

Figure 32

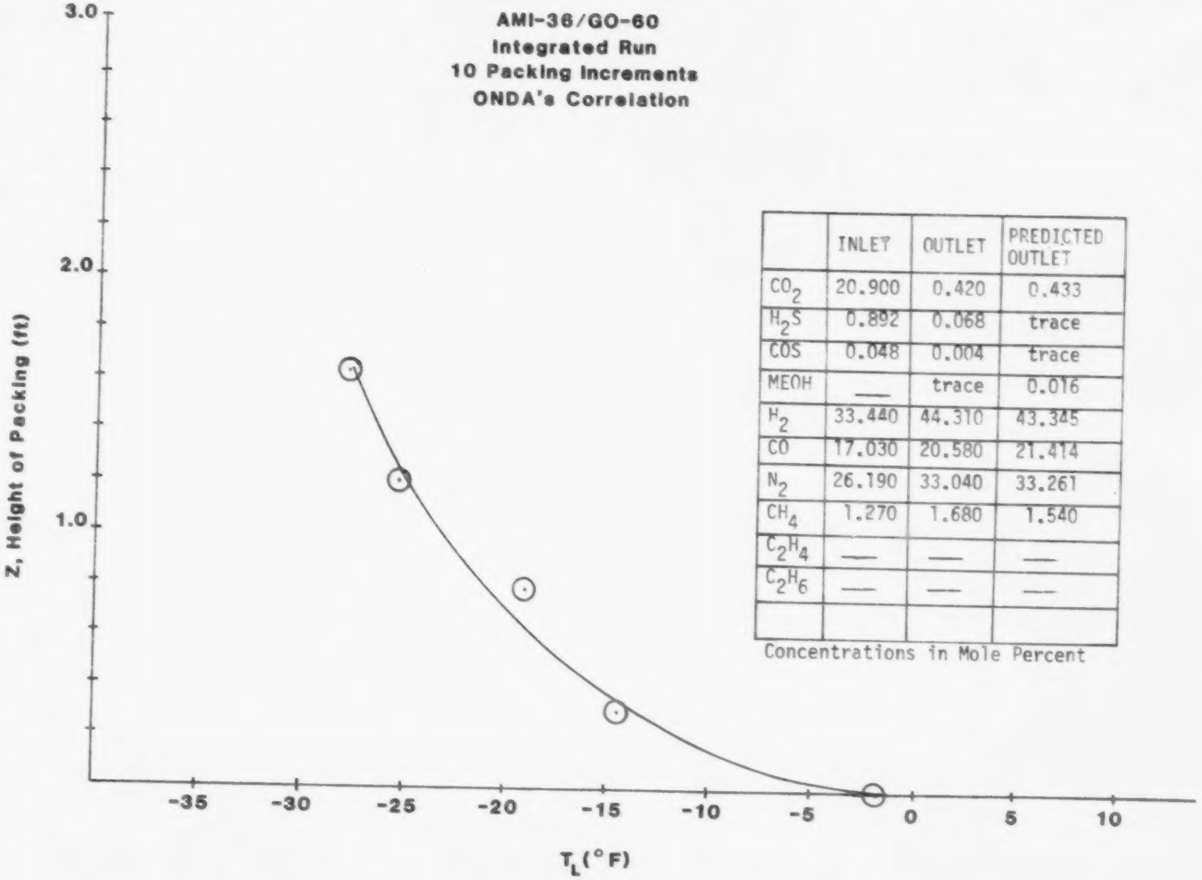
RUN NUMBER A-M-36
INTEGRATED RUN
DATE 7/18/1980

COLUMN TEMPERATURE PROFILE


SWEET GAS
10.63 SCFM
MEOH FLOW
0.790 GPM
-32.38 F
7.10 FT
-30.20 F
4.79 FT
ABSORBER PRESSURE =296.9 PSIG
TOTAL PACKING HEIGHT= 7.10 FEET
PACKING: 1/4" INTALOX SADDLES
-30.20 F
2.46 FT
-25.22 F
1.21 FT
-18.92 F
0.79 FT
-14.41 F
0.31 FT
0.00 FT
SOUR GAS INLET
13.63 SCFM
57.50 F
-1.72 F

	SOUR GAS (MOL %)	SWEET GAS (MOL %)
CO2	20.9000	0.4200
H2S	0.8918	0.0682
COS	0.0476	0.0038
MEOH	0.0000	0.0000
H2	33.4400	44.3100
CO	17.0300	20.5800
N2	26.1900	33.0400
CH4	1.2700	1.6800
C2H4	0.0000	0.0000
C2H6	0.0000	0.0000

Figure 33


AMI-36/GO-60
Integrated Run
10 Packing Increments
ONDA's Correlation

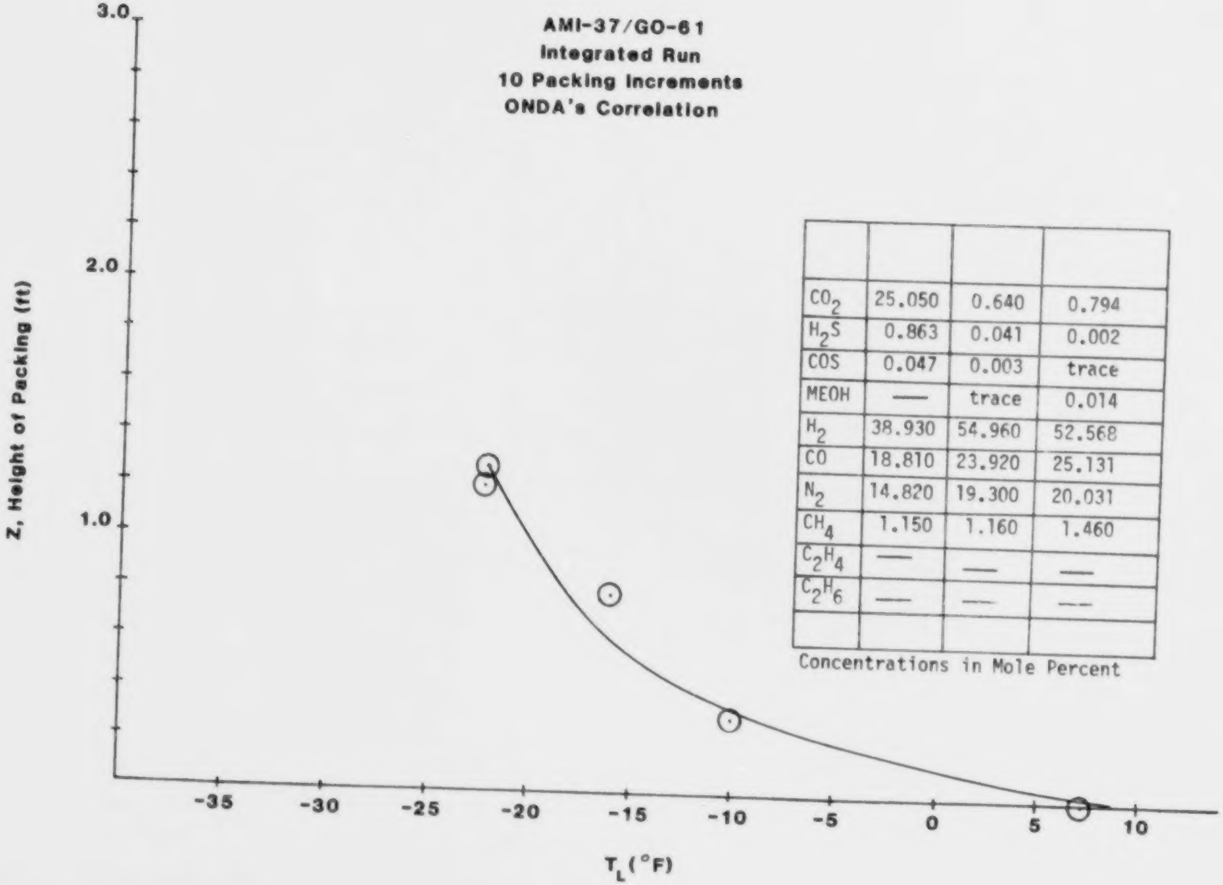
	INLET	OUTLET	PREDICTED OUTLET
CO_2	20.900	0.420	0.433
H_2S	0.892	0.068	trace
COS	0.048	0.004	trace
MEOH	—	trace	0.016
H_2	33.440	44.310	43.345
CO	17.030	20.580	21.414
N_2	26.190	33.040	33.261
CH_4	1.270	1.680	1.540
C_2H_4	—	—	—
C_2H_6	—	—	—

Concentrations in Mole Percent

Figure 34

```
                              RUN NUMBER A-M-37
                               INTEGRATED RUN
                              DATE    7/25/1980

                           COLUMN TEMPERATURE PROFILE
                           ------ ----------- -------

                                   .--------> SWEET GAS
                                   :          10.07 SCFM
                                   :
                                   :
                                   :
                              ********************
             MEOH FLOW        *                  *
              0.784 GPM       *                  *
             -36.33 F         *                  *
           ------------------>*                  *
                              *                  *
                              *==================*----     7.10 FT
                              *                  *
                              *                  *
                              *                  *
                              *                  *
                              *                  *
                              *                  *
                              *     -26.64 F     *----     4.79 FT
ABSORBER PRESSURE =446.6 PSIG *                  *
                              *                  *
TOTAL PACKING HEIGHT= 7.10 FEET *                *
                              *                  *
PACKING: 1/4" INTALOX SADDLES *                  *
                              *                  *
                              *     -27.54 F     *----     2.46 FT
                              *                  *
                              *                  *
                              *     -22.20 F     *----     1.21 FT
                              *                  *
                              *                  *
                              *     -16.03 F     *----     0.79 FT
                              *                  *
                              *                  *
                              *     -10.07 F     *----     0.31 FT
                              *                  *
                              *==================*----     0.00 FT
                              *                  *
         -------------------->*                  *
           SOUR GAS INLET     *                  *
           13.84 SCFM         *                  *
           59.92 F            *      7.19 F      *
                              *                  *-------------->
                              ********************

                             SOUR GAS (MOL %)        SWEET GAS (MOL %)
                             ---- ---                ----- ---

              CO2                25.0500                 0.6400
              H2S                 0.8631                 0.0405
              COS                 0.0475                 0.0027
              MEOH                0.0000                 0.0000
              H2                 38.9300                54.9600
              CO                 18.8100                23.9200
              N2                 14.8200                19.3000
              CH4                 1.1500                 1.1600
              C2H4                0.0000                 0.0000
              C2H6                0.0000                 0.0000
```

Figure 35



AMI-37/GO-61
Integrated Run
10 Packing Increments
ONDA's Correlation

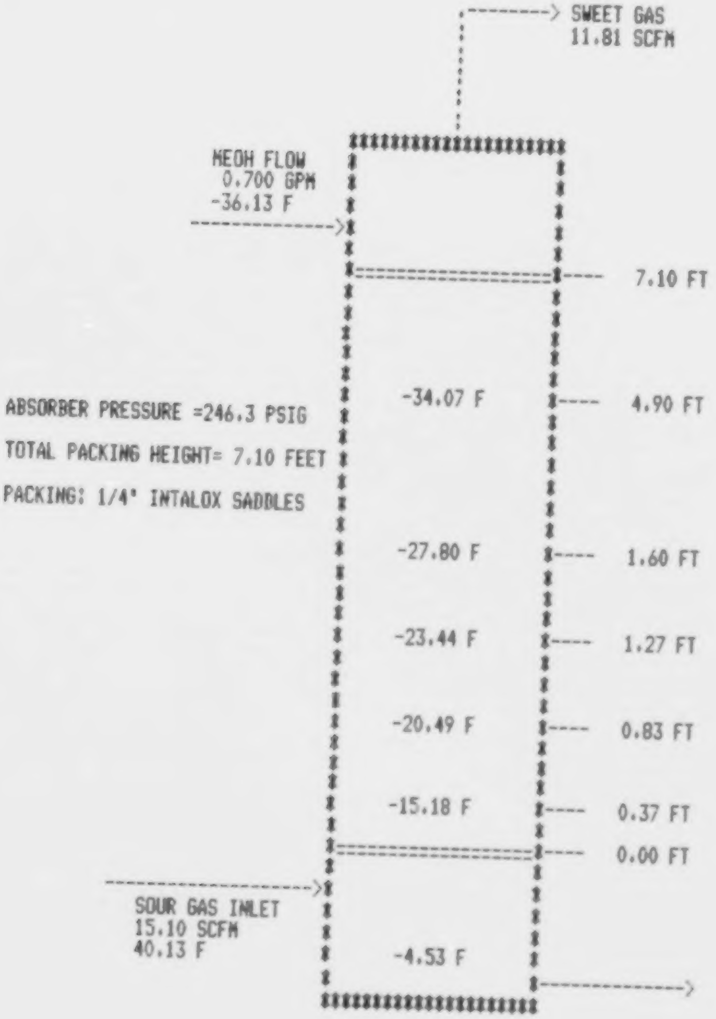
CO_2	25.050	0.640	0.794
H_2S	0.863	0.041	0.002
COS	0.047	0.003	trace
MEOH	—	trace	0.014
H_2	38.930	54.960	52.568
CO	18.810	23.920	25.131
N_2	14.820	19.300	20.031
CH_4	1.150	1.160	1.460
C_2H_4	—	—	—
C_2H_6	—	—	—

Concentrations in Mole Percent

Figure 36

RUN NUMBER A-M-43
INTEGRATED RUN
DATE 11/19/1980

COLUMN TEMPERATURE PROFILE

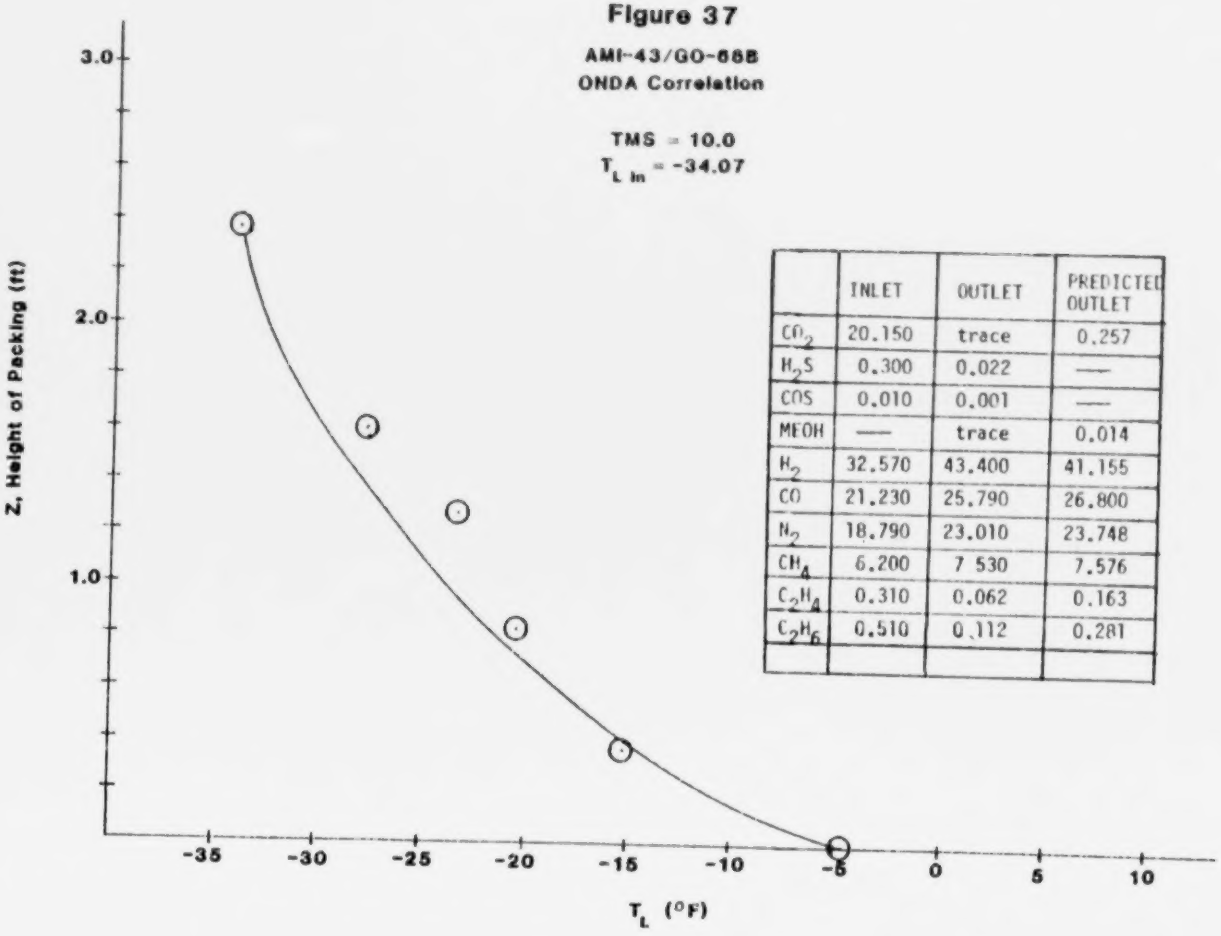

SWEET GAS
11.81 SCFM
MEOH FLOW
0.700 GPM
-36.13 F
7.10 FT
-34.07 F
4.90 FT
ABSORBER PRESSURE =246.3 PSIG
TOTAL PACKING HEIGHT= 7.10 FEET
PACKING: 1/4" INTALOX SADDLES
-27.80 F
1.60 FT
-23.44 F
1.27 FT
-20.49 F
0.83 FT
-15.18 F
0.37 FT
0.00 FT
SOUR GAS INLET
15.10 SCFM
40.13 F
-4.53 F

	SOUR GAS (MOL %)	SWEET GAS (MOL %)
CO2	20.0500	0.0000
H2S	0.2950	0.0219
COS	0.0120	0.0012
MEOH	0.0000	0.0000
H2	32.4200	43.4000
CO	21.2300	25.7900
N2	18.7000	23.0100
CH4	6.5900	7.5300
C2H4	0.3080	0.0617
C2H6	0.5080	0.1124

Figure 37

AMI-43/GO-68B
ONDA Correlation

TMS = 10.0
$T_{L\,in} = -34.07$

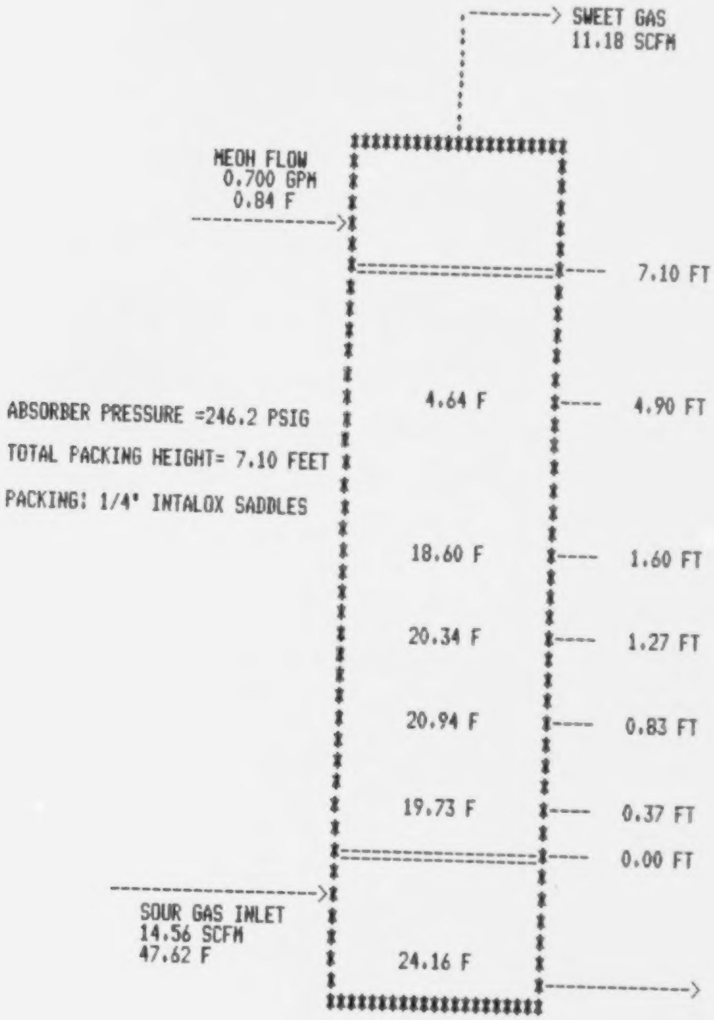


	INLET	OUTLET	PREDICTED OUTLET
CO_2	20.150	trace	0.257
H_2S	0.300	0.022	——
COS	0.010	0.001	——
MEOH	——	trace	0.014
H_2	32.570	43.400	41.155
CO	21.230	25.790	26.800
N_2	18.790	23.010	23.748
CH_4	6.200	7 530	7.576
C_2H_4	0.310	0.062	0.163
C_2H_6	0.510	0.112	0.281

Figure 38

RUN NUMBER A-M-44
INTEGRATED RUN
DATE 12/15/1980

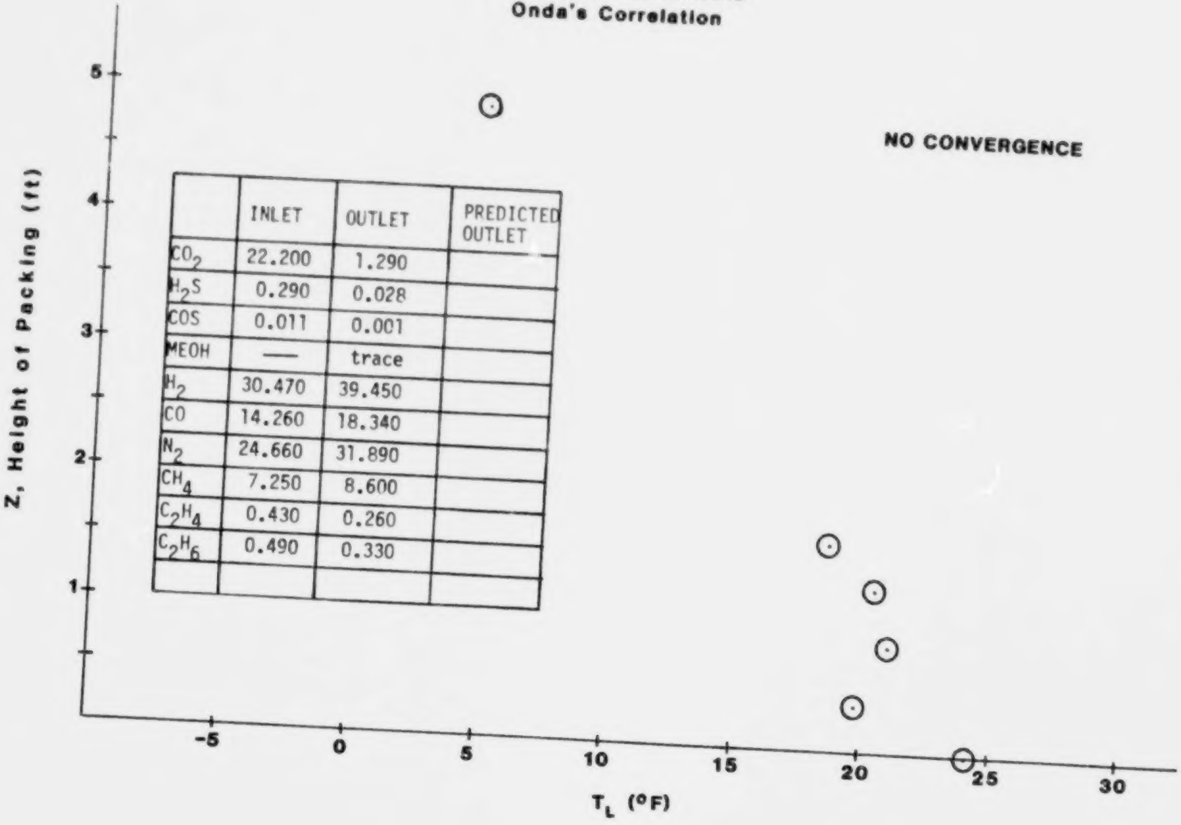
COLUMN TEMPERATURE PROFILE


SWEET GAS
11.18 SCFM
MEOH FLOW
0.700 GPM
0.84 F
7.10 FT
4.64 F
4.90 FT
ABSORBER PRESSURE =246.2 PSIG
TOTAL PACKING HEIGHT= 7.10 FEET
PACKING: 1/4" INTALOX SADDLES
18.60 F
1.60 FT
20.34 F
1.27 FT
20.94 F
0.83 FT
19.73 F
0.37 FT
0.00 FT
SOUR GAS INLET
14.56 SCFM
47.62 F
24.16 F

	SOUR GAS (MOL %)	SWEET GAS (MOL %)
CO2	22.2000	1.2900
H2S	0.2900	0.0280
COS	0.0112	0.0010
MEOH	0.0000	0.0000
H2	30.4700	39.4500
CO	14.2600	18.3400
N2	24.6600	31.8900
CH4	7.2500	8.6000
C2H4	0.4300	0.2600
C2H6	0.4900	0.3300

Figure 39

AMI-44/GO-69B
Integrated Run
10 Packing Increments
Onda's Correlation

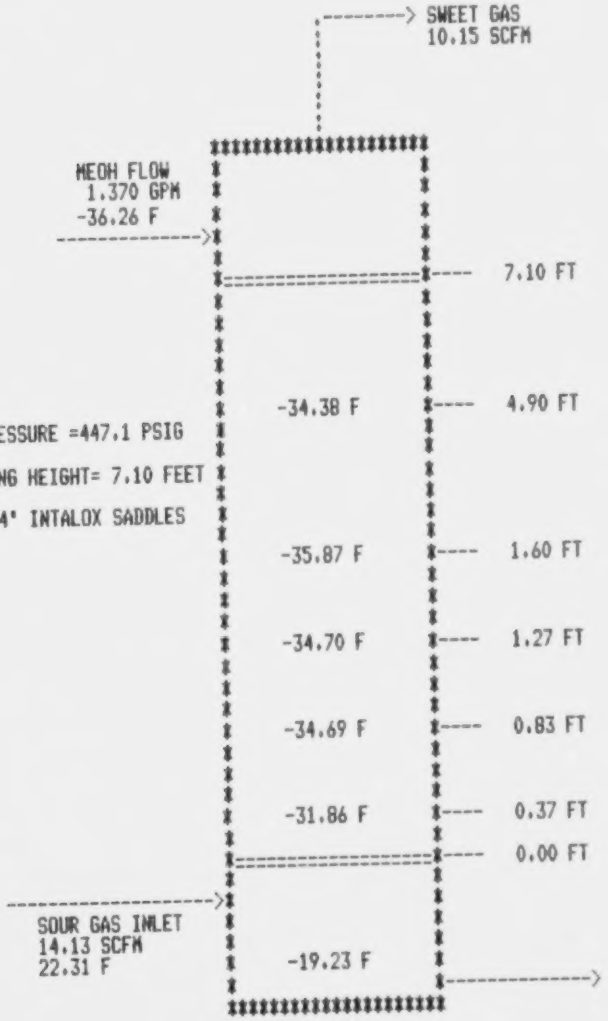


	INLET	OUTLET	PREDICTED OUTLET
CO_2	22.200	1.290	
H_2S	0.290	0.028	
COS	0.011	0.001	
MEOH	——	trace	
H_2	30.470	39.450	
CO	14.260	18.340	
N_2	24.660	31.890	
CH_4	7.250	8.600	
C_2H_4	0.430	0.260	
C_2H_6	0.490	0.330	

Figure 40

RUN NUMBER A-M-45
INTEGRATED RUN
DATE 1/15/1981

COLUMN TEMPERATURE PROFILE


SWEET GAS
10.15 SCFM
MEOH FLOW
1.370 GPM
-36.26 F
7.10 FT
-34.38 F
4.90 FT
-35.87 F
1.60 FT
-34.70 F
1.27 FT
-34.69 F
0.83 FT
-31.86 F
0.37 FT
0.00 FT
SOUR GAS INLET
14.13 SCFM
22.31 F
-19.23 F

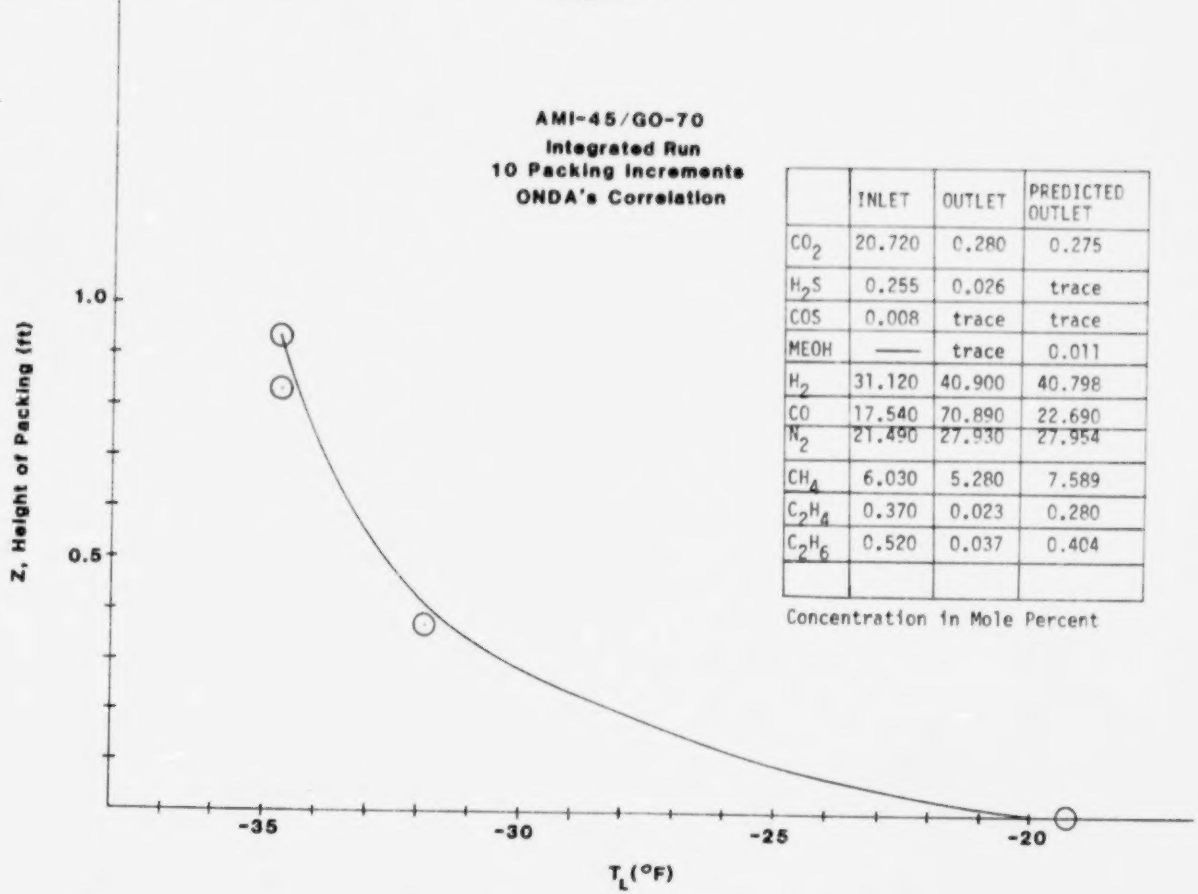
ABSORBER PRESSURE =447.1 PSIG

TOTAL PACKING HEIGHT= 7.10 FEET

PACKING: 1/4" INTALOX SADDLES

	SOUR GAS (MOL %)	SWEET GAS (MOL %)
CO2	20.7200	2.5800
H2S	0.2550	0.0260
COS	0.0079	0.0005
MEOH	0.0000	0.0000
H2	31.1200	40.9000
CO	17.5400	20.8900
N2	21.4900	27.9300
CH4	6.0300	5.2800
C2H4	0.3700	0.0230
C2H6	0.5200	0.0370

Figure 41



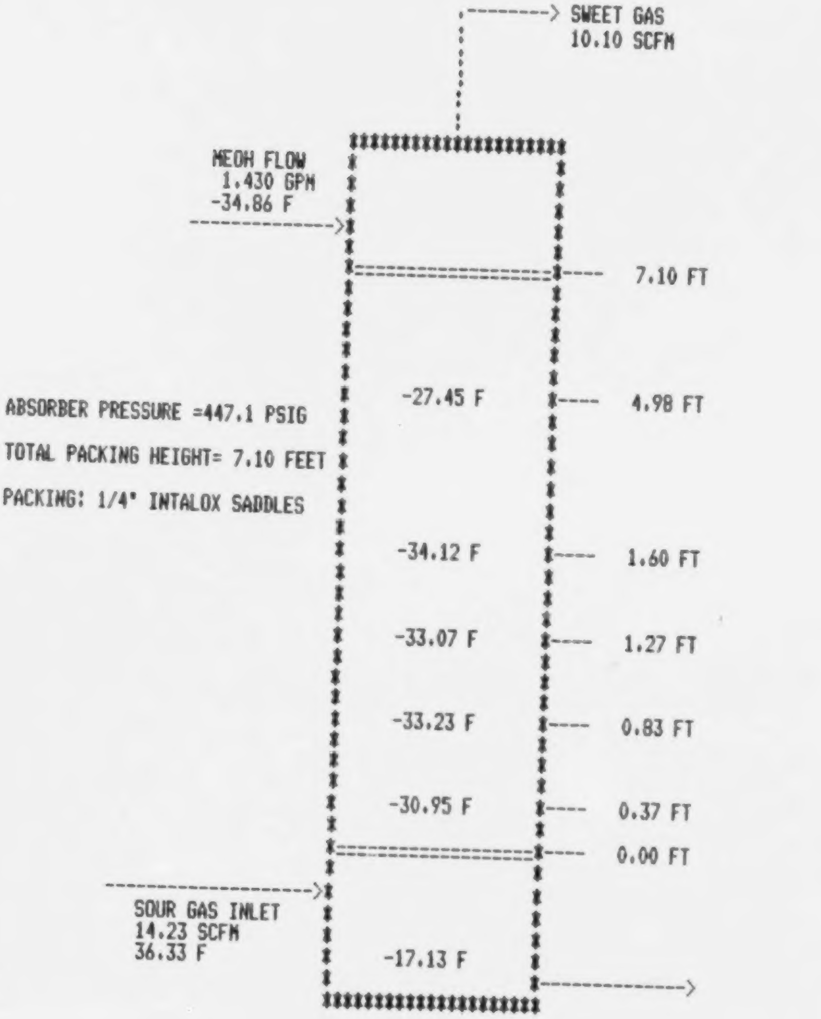
	INLET	OUTLET	PREDICTED OUTLET
CO_2	20.720	0.280	0.275
H_2S	0.255	0.026	trace
COS	0.008	trace	trace
MEOH	——	trace	0.011
H_2	31.120	40.900	40.798
CO	17.540	70.890	22.690
N_2	21.490	27.930	27.954
CH_4	6.030	5.280	7.589
C_2H_4	0.370	0.023	0.280
C_2H_6	0.520	0.037	0.404

Concentration in Mole Percent

Figure 42

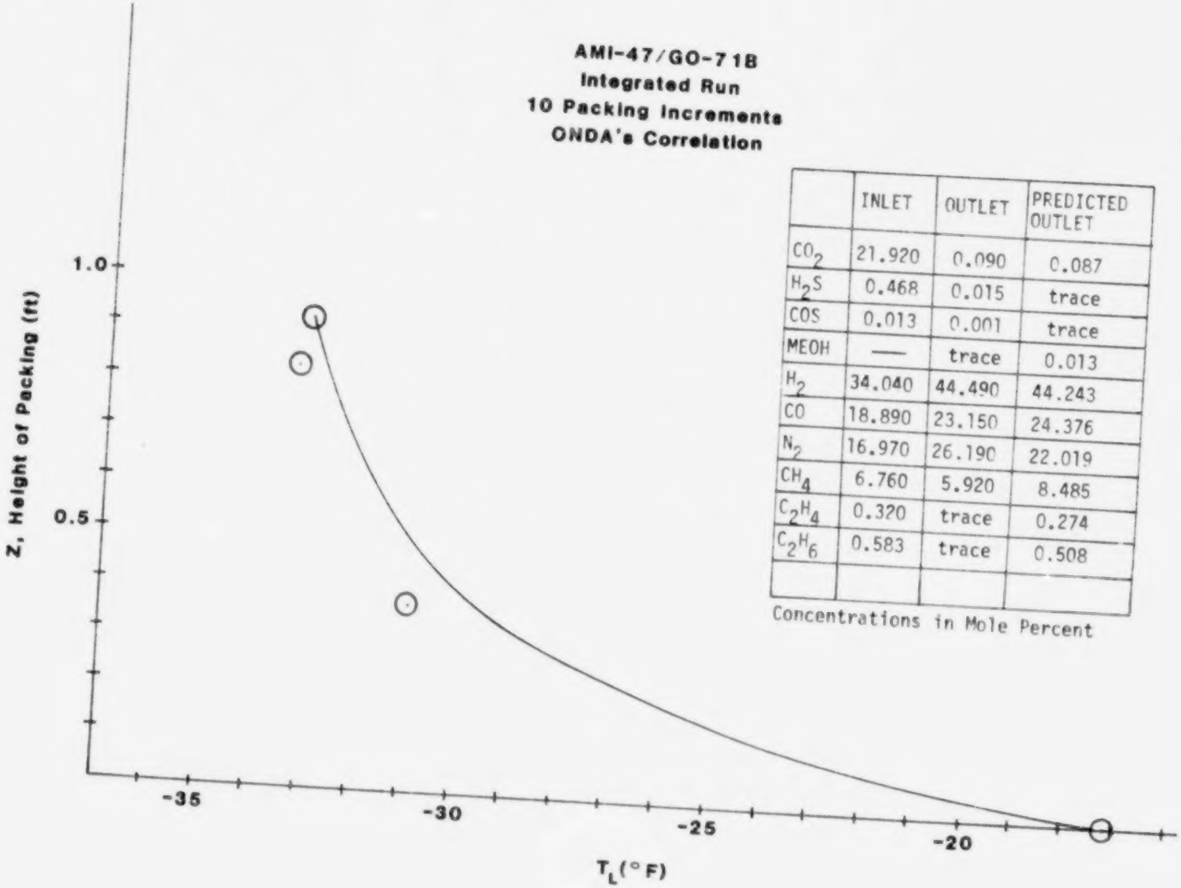
RUN NUMBER A-M-47
INTEGRATED
DATE 1/27/1981

COLUMN TEMPERATURE PROFILE


SWEET GAS
10.10 SCFM
MEOH FLOW
1.430 GPM
-34.86 F
7.10 FT
-27.45 F
4.98 FT
ABSORBER PRESSURE =447.1 PSIG
TOTAL PACKING HEIGHT= 7.10 FEET
PACKING: 1/4" INTALOX SADDLES
-34.12 F
1.60 FT
-33.07 F
1.27 FT
-33.23 F
0.83 FT
-30.95 F
0.37 FT
0.00 FT
SOUR GAS INLET
14.23 SCFM
36.33 F
-17.13 F

	SOUR GAS (MOL %)	SWEET GAS (MOL %)
CO2	21.9200	0.0900
H2S	0.4682	0.0151
COS	0.0133	0.0007
MEOH	0.0000	0.0000
H2	34.0400	44.9900
CO	18.8900	23.1500
N2	16.9700	26.1900
CH4	6.7600	5.9200
C2H4	0.3197	0.0000
C2H6	0.5827	0.0000

Figure 43



AMI-47/GO-71B
Integrated Run
10 Packing Increments
ONDA's Correlation

	INLET	OUTLET	PREDICTED OUTLET
CO_2	21.920	0.090	0.087
H_2S	0.468	0.015	trace
COS	0.013	0.001	trace
MEOH	——	trace	0.013
H_2	34.040	44.490	44.243
CO	18.890	23.150	24.376
N_2	16.970	26.190	22.019
CH_4	6.760	5.920	8.485
C_2H_4	0.320	trace	0.274
C_2H_6	0.583	trace	0.508

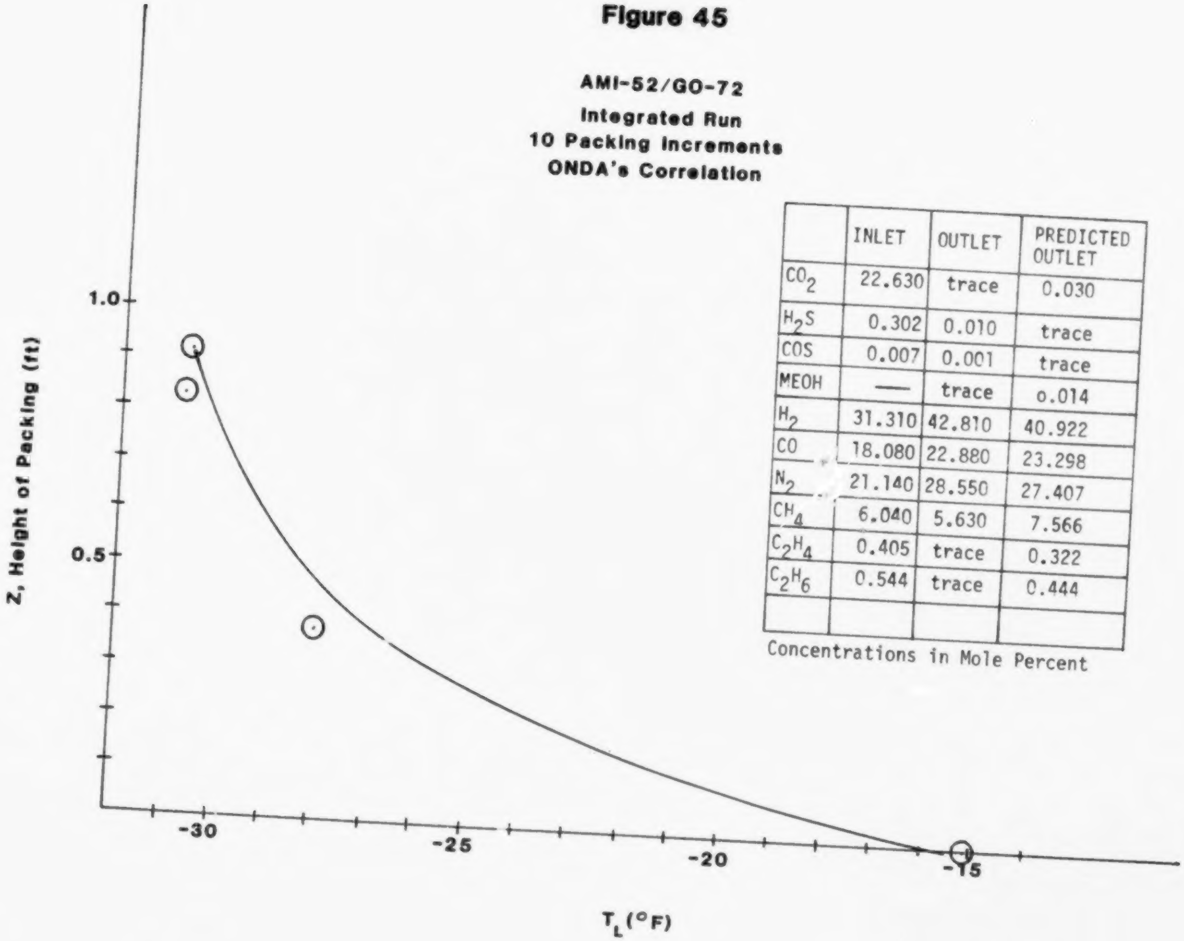
Concentrations in Mole Percent

Figure 44

```
                     RUN NUMBER A-M-52
                       INTEGRATED
                      DATE    2/19/1981

                COLUMN TEMPERATURE PROFILE
                ------ ----------- -------

                             .--------> SWEET GAS
                             :            9.70 SCFM
                             :
                             :
                             :
                  ***********************
      MEOH FLOW   *                     *
       1.400 GPM  *                     *
      -35.66 F    *                     *
    ------------->*                     *
                  *=====================*----    7.10 FT
                  *                     *
                  *                     *
                  *                     *
                  *       -32.11 F      *----    4.98 FT
ABSORBER PRESSURE =447.3 PSIG           *
                  *                     *
TOTAL PACKING HEIGHT= 7.10 FEET         *
                  *                     *
PACKING: 1/4" INTALOX SADDLES           *
                  *                     *
                  *       -31.70 F      *----    1.60 FT
                  *                     *
                  *       -30.54 F      *----    1.27 FT
                  *                     *
                  *       -30.71 F      *----    0.83 FT
                  *                     *
                  *       -28.00 F      *----    0.37 FT
                  *=====================*----    0.00 FT
    ------------->*                     *
      SOUR GAS INLET                    *
      14.00 SCFM  *                     *
      38.05 F     *       -15.11 F      *
                  *                     *-------------->
                  ***********************

                     SOUR GAS (MOL %)         SWEET GAS (MOL %)
                     ---- ---                 ----- ---

      CO2                22.6300                   0.0000
      H2S                 0.3023                   0.0105
      COS                 0.0067                   0.0006
      MEOH                0.0000                   0.0000
      H2                 31.3100                  42.8100
      CO                 18.0800                  22.8800
      N2                 21.1400                  28.5500
      CH4                 6.0400                   5.6300
      C2H4                0.4048                   0.0000
      C2H6                0.5440                   0.0000
```

Figure 45

AMI-52/GO-72
Integrated Run
10 Packing Increments
ONDA's Correlation

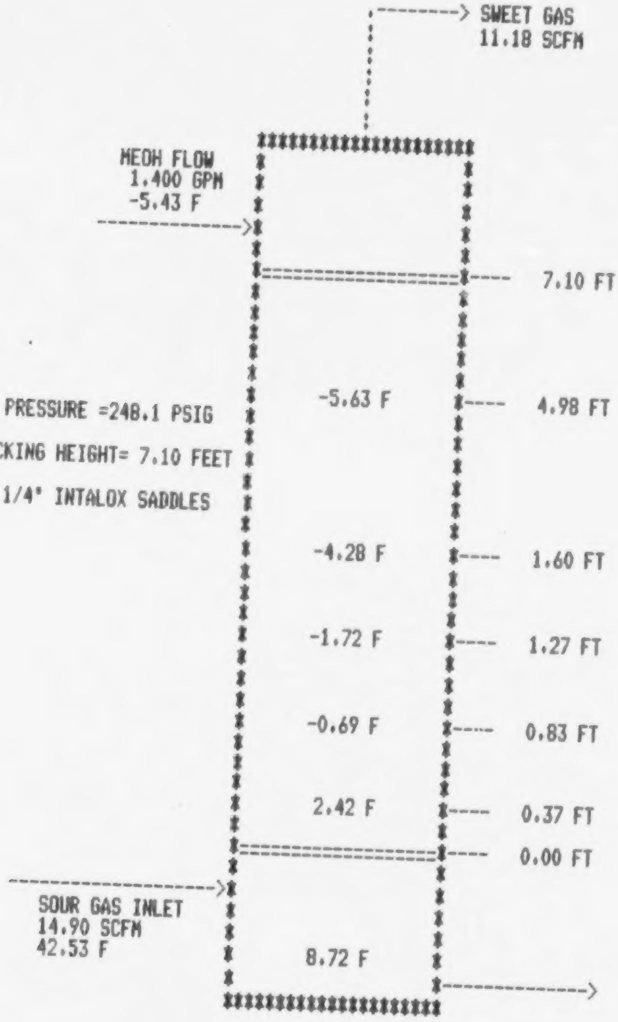


	INLET	OUTLET	PREDICTED OUTLET
CO_2	22.630	trace	0.030
H_2S	0.302	0.010	trace
COS	0.007	0.001	trace
MEOH	—	trace	o.014
H_2	31.310	42.810	40.922
CO	18.080	22.880	23.298
N_2	21.140	28.550	27.407
CH_4	6.040	5.630	7.566
C_2H_4	0.405	trace	0.322
C_2H_6	0.544	trace	0.444

Concentrations in Mole Percent

Figure 46

```
RUN NUMBER A-M-53
 INTEGRATED
DATE    3/12/1981

COLUMN TEMPERATURE PROFILE
------ ----------- -------
```


SWEET GAS
11.18 SCFM
MEOH FLOW
1.400 GPM
-5.43 F
7.10 FT
-5.63 F
4.98 FT
-4.28 F
1.60 FT
-1.72 F
1.27 FT
-0.69 F
0.83 FT
2.42 F
0.37 FT
0.00 FT
SOUR GAS INLET
14.90 SCFM
42.53 F
8.72 F

ABSORBER PRESSURE =248.1 PSIG

TOTAL PACKING HEIGHT= 7.10 FEET

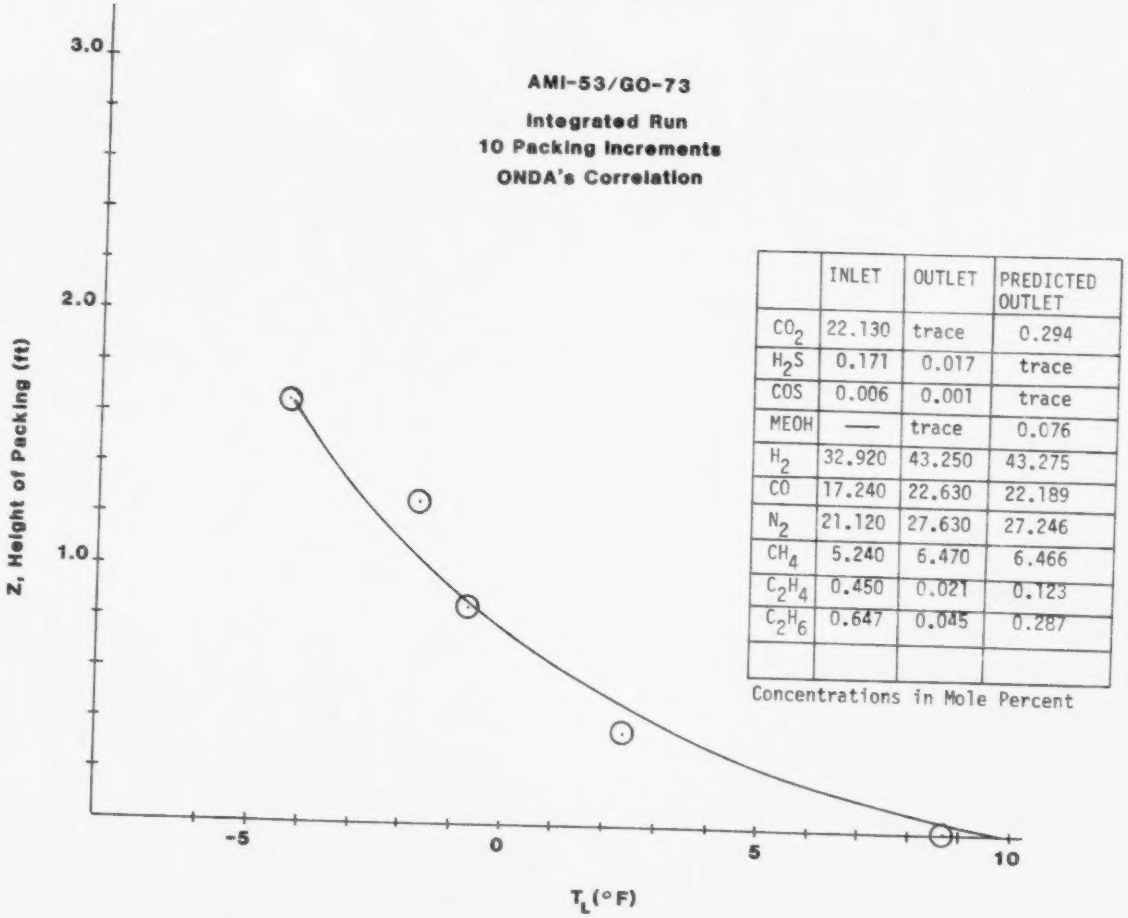
PACKING: 1/4" INTALOX SADDLES

```
            SOUR GAS (MOL %)      SWEET GAS (MOL %)
            ---- ---              ----- ---

CO2            22.1300               0.0000
H2S             0.1710               0.0172
COS             0.0060               0.0007
MEOH            0.0000               0.0000
H2             32.9200              43.2500
CO             17.2400              22.6300
N2             21.1200              27.6300
CH4             5.2900               6.4700
C2H4            0.4497               0.0212
C2H6            0.6470               0.0454
```

Figure 47

AMI-53/GO-73

Integrated Run
10 Packing Increments
ONDA's Correlation



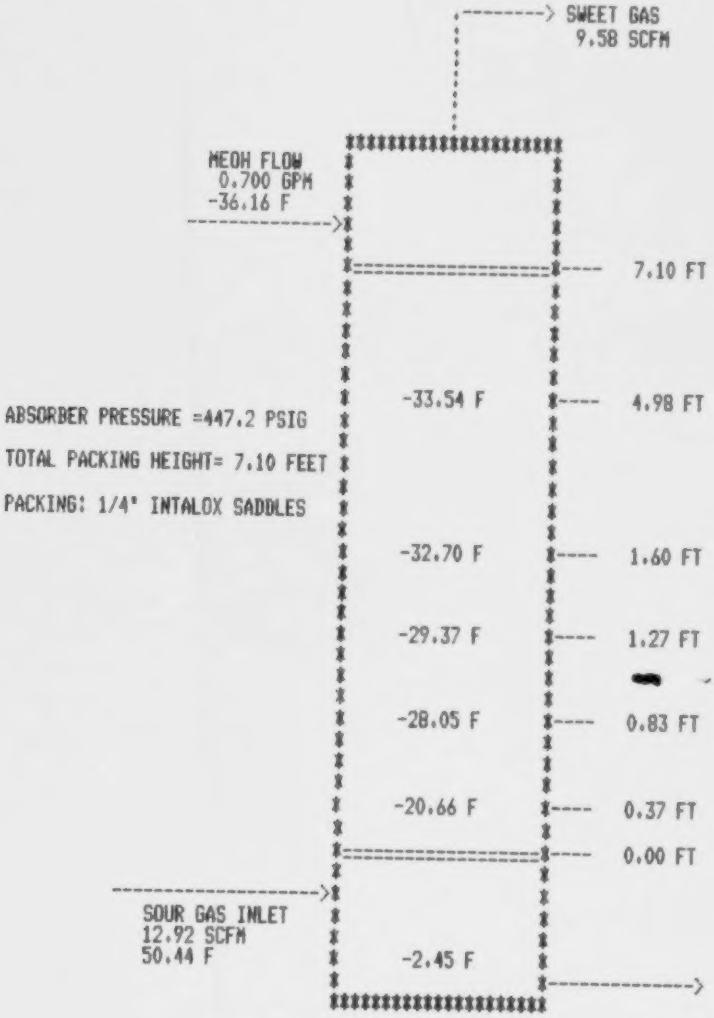
	INLET	OUTLET	PREDICTED OUTLET
CO_2	22.130	trace	0.294
H_2S	0.171	0.017	trace
COS	0.006	0.001	trace
MEOH	—	trace	0.076
H_2	32.920	43.250	43.275
CO	17.240	22.630	22.189
N_2	21.120	27.630	27.246
CH_4	5.240	6.470	6.466
C_2H_4	0.450	0.021	0.123
C_2H_6	0.647	0.045	0.287

Concentrations in Mole Percent

Figure 48

RUN NUMBER A-M-57
INTEGRATED RUN
DATE 4/28/1981

COLUMN TEMPERATURE PROFILE


SWEET GAS
9.58 SCFM
MEOH FLOW
0.700 GPM
-36.16 F
7.10 FT
-33.54 F
4.98 FT
-32.70 F
1.60 FT
-29.37 F
1.27 FT
-28.05 F
0.83 FT
-20.66 F
0.37 FT
0.00 FT
SOUR GAS INLET
12.92 SCFM
50.44 F
-2.45 F

ABSORBER PRESSURE =447.2 PSIG

TOTAL PACKING HEIGHT= 7.10 FEET

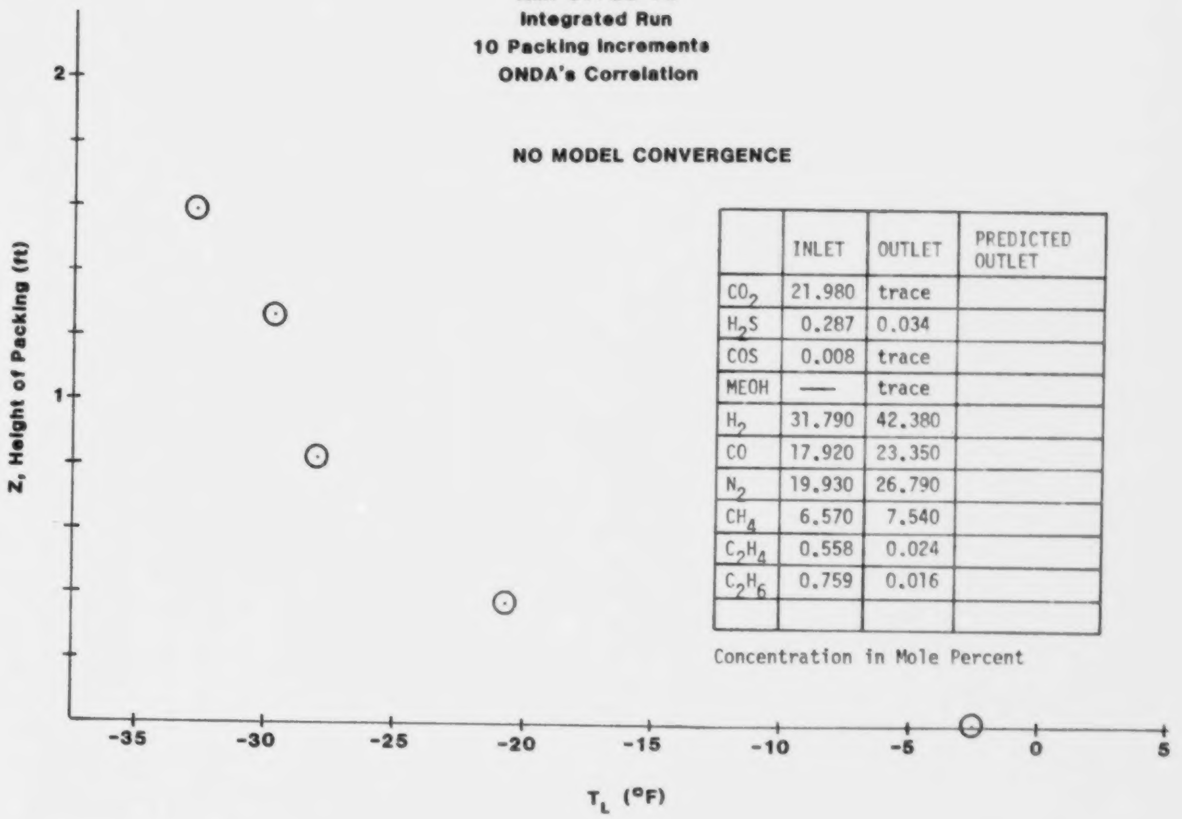
PACKING: 1/4" INTALOX SADDLES

	SOUR GAS (MOL %)	SWEET GAS (MOL %)
CO2	21.9800	0.0000
H2S	0.2867	0.0034
COS	0.0078	0.0000
MEOH	0.0000	0.0000
H2	31.7900	42.3800
CO	17.9200	23.3500
N2	19.9300	26.7900
CH4	6.5700	7.5400
C2H4	0.5577	0.0242
C2H6	0.7588	0.0164

Figure 49

AMI-57/GO-78
Integrated Run
10 Packing Increments
ONDA's Correlation

NO MODEL CONVERGENCE



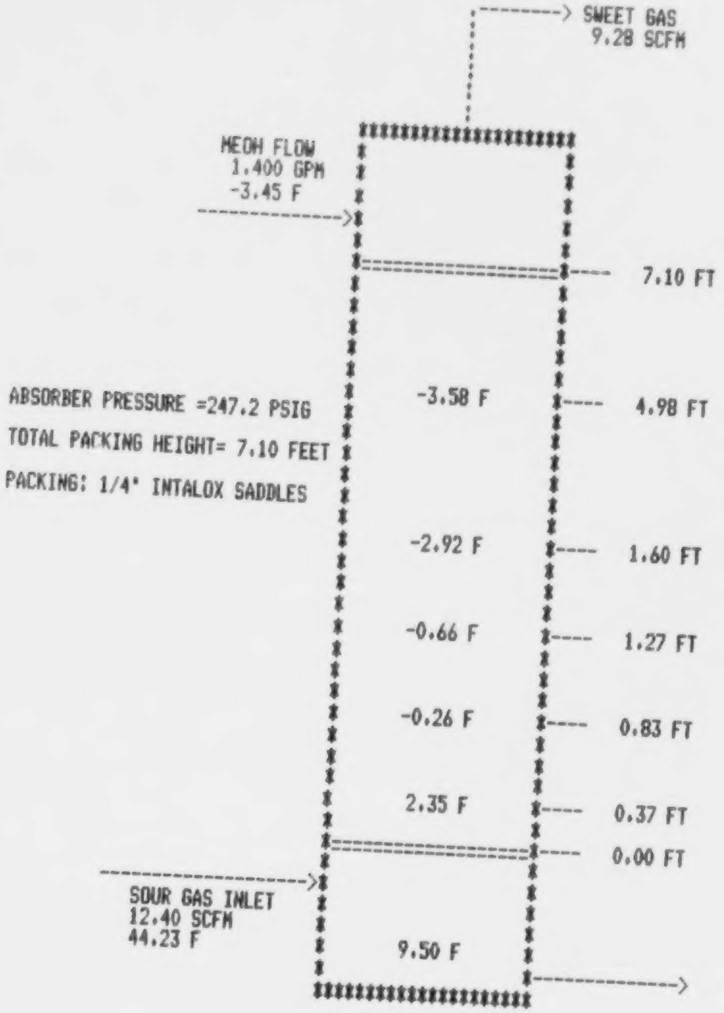
	INLET	OUTLET	PREDICTED OUTLET
CO_2	21.980	trace	
H_2S	0.287	0.034	
COS	0.008	trace	
MEOH	——	trace	
H_2	31.790	42.380	
CO	17.920	23.350	
N_2	19.930	26.790	
CH_4	6.570	7.540	
C_2H_4	0.558	0.024	
C_2H_6	0.759	0.016	

Concentration in Mole Percent

Figure 50

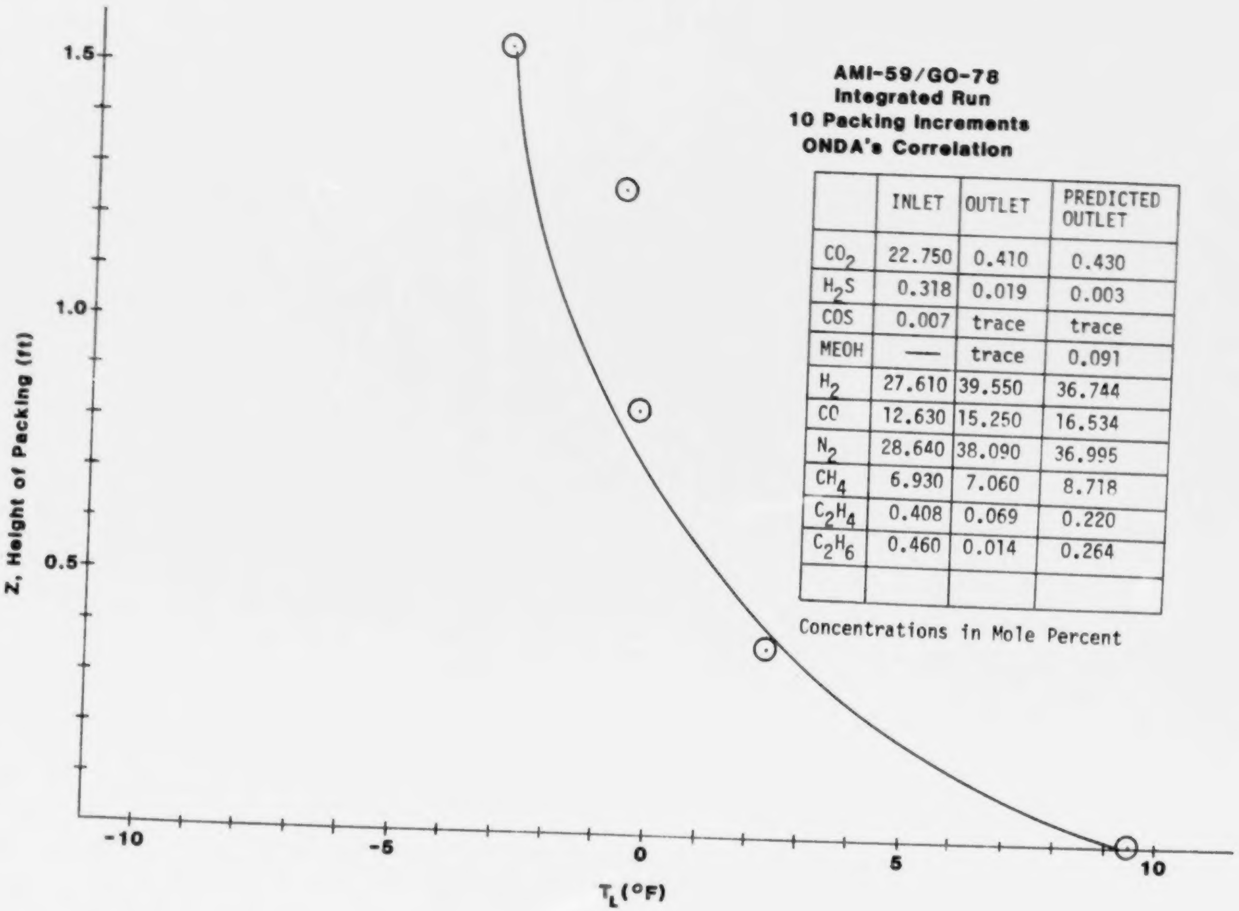
RUN NUMBER A-M-59
INTEGRATED RUN
DATE 5/25/1981

COLUMN TEMPERATURE PROFILE


SWEET GAS
9.28 SCFM
MEOH FLOW
1.400 GPM
-3.45 F
7.10 FT
ABSORBER PRESSURE =247.2 PSIG
TOTAL PACKING HEIGHT= 7.10 FEET
PACKING: 1/4" INTALOX SADDLES
-3.58 F
4.98 FT
-2.92 F
1.60 FT
-0.66 F
1.27 FT
-0.26 F
0.83 FT
2.35 F
0.37 FT
0.00 FT
SOUR GAS INLET
12.40 SCFM
44.23 F
9.50 F

	SOUR GAS (MOL %)	SWEET GAS (MOL %)
CO2	22.7500	0.4100
H2S	0.3180	0.0190
COS	0.0069	0.0004
MEOH	0.0000	0.0000
H2	27.6100	39.5500
CO	12.6300	15.2500
N2	28.6400	38.0900
CH4	6.9300	7.0600
C2H4	0.4080	0.0690
C2H6	0.4600	0.0140

Figure 51



AMI-59/GO-78
Integrated Run
10 Packing Increments
ONDA's Correlation

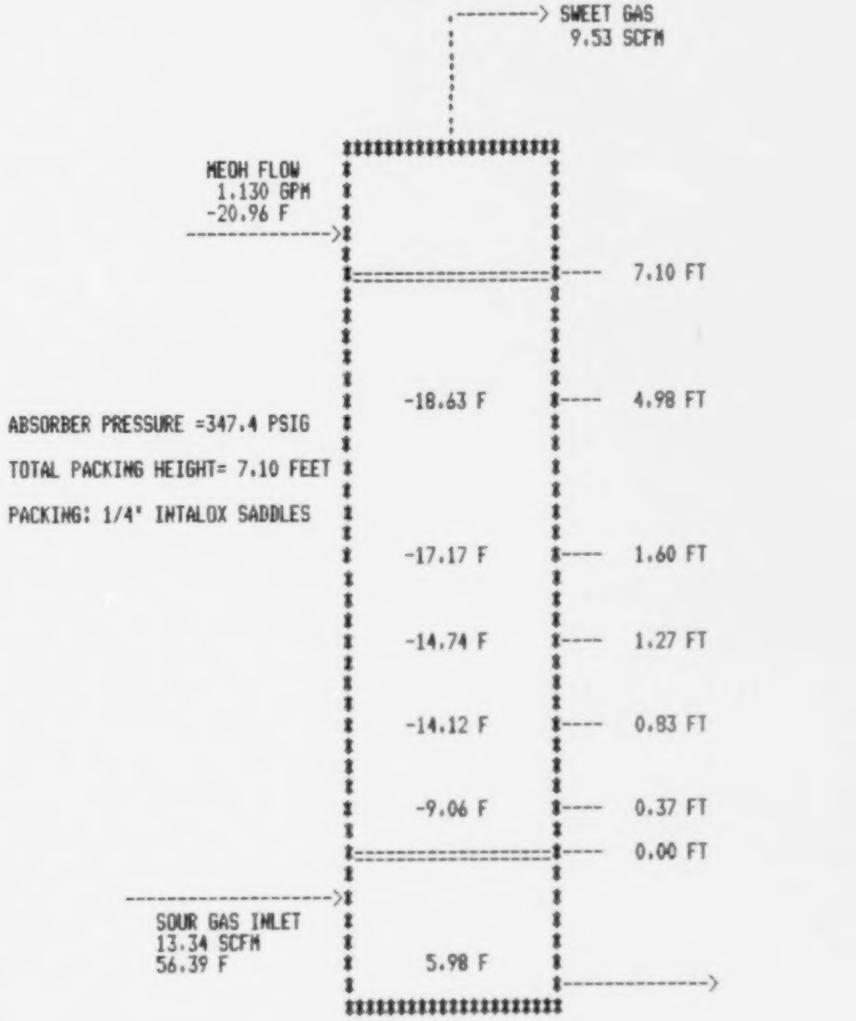
	INLET	OUTLET	PREDICTED OUTLET
CO_2	22.750	0.410	0.430
H_2S	0.318	0.019	0.003
COS	0.007	trace	trace
MEOH	—	trace	0.091
H_2	27.610	39.550	36.744
CO	12.630	15.250	16.534
N_2	28.640	38.090	36.995
CH_4	6.930	7.060	8.718
C_2H_4	0.408	0.069	0.220
C_2H_6	0.460	0.014	0.264

Concentrations in Mole Percent

Figure 52

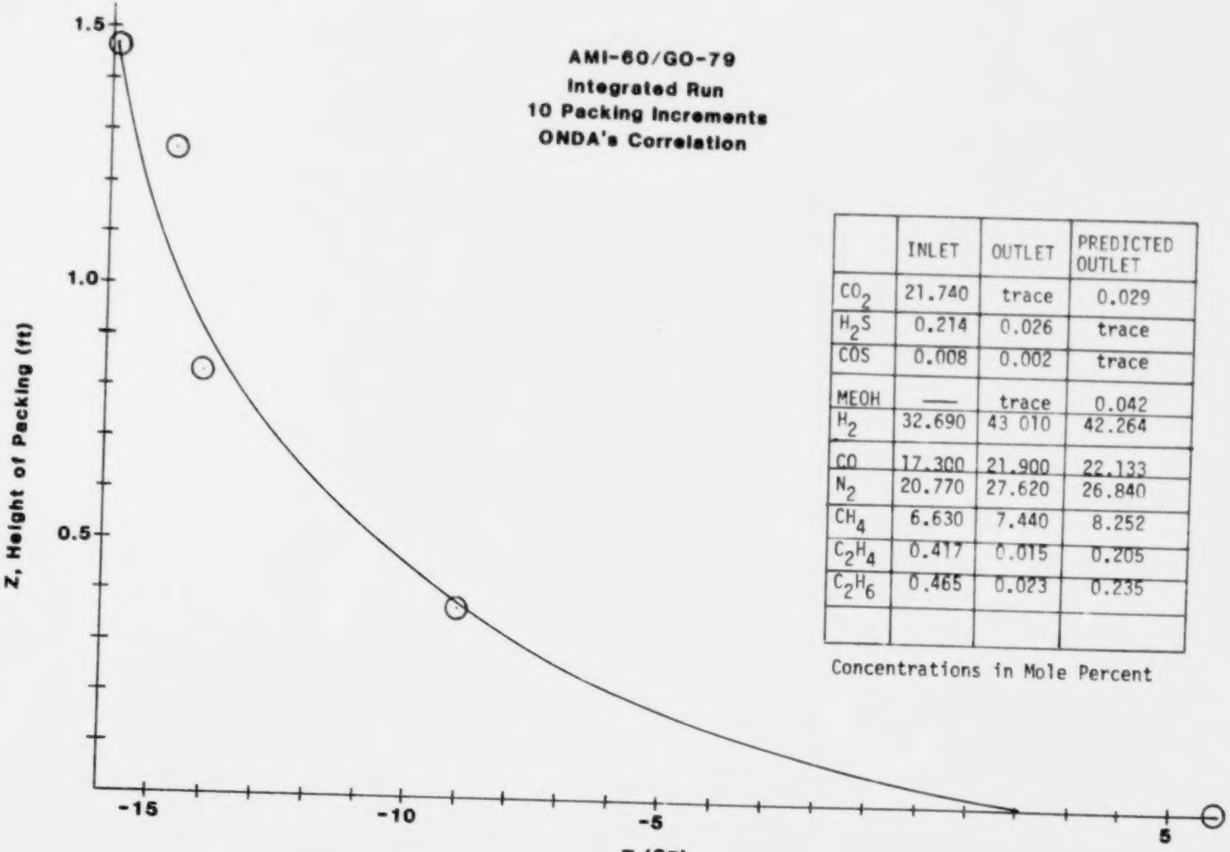
RUN NUMBER A-M-60
INTEGRATED RUN
DATE 6/10/1981

COLUMN TEMPERATURE PROFILE


SWEET GAS
9.53 SCFM
MEOH FLOW
1.130 GPM
-20.96 F
7.10 FT
-18.63 F
4.98 FT
ABSORBER PRESSURE =347.4 PSIG
TOTAL PACKING HEIGHT= 7.10 FEET
PACKING: 1/4" INTALOX SADDLES
-17.17 F
1.60 FT
-14.74 F
1.27 FT
-14.12 F
0.83 FT
-9.06 F
0.37 FT
0.00 FT
SOUR GAS INLET
13.34 SCFM
56.39 F
5.98 F

	SOUR GAS (MOL %)	SWEET GAS (MOL %)
CO2	21.7400	0.0000
H2S	0.2139	0.0260
COS	0.0084	0.0019
MEOH	0.0000	0.0000
H2	32.6900	43.0100
CO	17.3000	21.9000
N2	20.7700	27.6200
CH4	6.6300	7.4400
C2H4	0.4174	0.0148
C2H6	0.4652	0.0233

Figure 53



AMI-60/GO-79
Integrated Run
10 Packing Increments
ONDA's Correlation

	INLET	OUTLET	PREDICTED OUTLET
CO_2	21.740	trace	0.029
H_2S	0.214	0.026	trace
COS	0.008	0.002	trace
MEOH	——	trace	0.042
H_2	32.690	43 010	42.264
CO	17.300	21.900	22.133
N_2	20.770	27.620	26.840
CH_4	6.630	7.440	8.252
C_2H_4	0.417	0.015	0.205
C_2H_6	0.465	0.023	0.235

Concentrations in Mole Percent

TABLE 9

INTEGRATED RUN RESULTS

RUN	L	T_L	G	T_G	P	CO_2 RECOVERY	COMPUTED HEIGHT OF PACKING (ONDA)
30	60.82	-28.27	16.21	54.04	400	0.958	1.416
35	60.82	-34.65	15.86	53.86	400	0.991	1.501
36	72.76	-27.75	16.39	57.50	300	0.984	1.640
37	71.42	-22.20	16.64	59.92	450	0.977	1.282
43	63.55	-34.07	18.08	40.13	250	0.990	2.681
44	63.51	0.84	16.96	47.62	250	0.950	NO CONVERGENCE
45	129.32	-34.07	16.99	22.31	450	0.990	0.937
47	130.48	-33.07	17.11	36.33	450	0.990	0.920
52	127.87	-30.70	16.83	38.05	450	0.999	0.988
53	127.46	- 4.28	17.88	42.53	250	0.999	2.009
57	61.66	-32.70	15.39	50.44	450	0.990	NO CONVERGENCE
59	127.30	- 2.92	14.91	44.23	250	0.986	1.543
60	100.80	-16.00	16.04	56.39	350	0.999	1.464

L	LB-MOLES/HR/FT^2
G	LB-MOLES/HR/FT^2
T_L	DEGREES F
T_G	DEGREES F
P	PSIG

HEIGHT OF PACKING IN FEET

TABLE 10

COMPARISON OF PREDICTED AND EXPERIMENTAL OUTLET LIQUID TEMPERATURES FOR INTEGRATED RUNS

ACRS RUN	GASIFIER RUN	MASS BALANCE	TL IN	TL OUT	COMPUTED TL OUT
30	56	100.0	-28.27	4.83	5.47
35	59	102.4	-34.65	0.72	3.11
36	60	104.0	-27.75	-1.72	- 0.84
37	61	103.3	-22.20	7.19	8.22
43	68B	103.8	-34.07	-4.53	- 2.82
44	69	102.3	0.84	24.16	-----
45	70	103.0	-34.07	-19.23	-19.74
47	71B	104.8	-33.07	-17.13	-17.35
52	72	101.4	-30.70	-15.11	-14.81
53	73	103.1	- 4.28	8.72	10.13
57	76	99.2	-32.70	- 2.45	-----
59	78	101.8	- 2.92	9.50	9.29
60	79	95.5	-16.00	5.98	2.44

WHERE ALL TEMPERATURES ARE DEGREES F

Note that runs AMI-44 and 57 failed to converge and are not included in the comparison of the predicted and experimental data. They are discussed further in the section on minimum liquid to gas ratios.

Discussion

For purpose of illustration, computer output from AMI-53 will be presented in its entirety. Table 11 lists the computer output from AMI-53 for the iteration in which the calculation converged.

TABLE 11

```
          Run AM-53   MCOMP Simulation   07/28/81   10:45

   Gas Mass Vel.    Gas Temp. In    Liq. Mass Vel.    Liq. Temp. In
   Mol/Hr/Sq.Ft       Deg. F        Mol/Hr/Sq.Ft        Deg. F

     17.88            42.53           127.46             -4.28

                 Pressure    The Key     Fract Key
                 Atmos.      Comp.     Comp. Absorber

                 17.890        1          0.99900

          Comp. No.    Comp.     Inlet Gas     Inlet Liquid
                                 Mol.Fract      Mol.Fract

              1        CO2        0.2213         0.0000
              2        H2S        0.0017         0.0000
              3        COS        0.0001         0.0000
              4        MEOH       0.0000         1.0000
              5        H2         0.3292         0.0000
              6        CO         0.1724         0.0000
              7        N2         0.2112         0.0000
              8        CH4        0.0529         0.0000
              9        C2H4       0.0045         0.0000
             10        C2H6       0.0065         0.0000
             11        C3H6       0.0000         0.0000
             12        C3H8       0.0000         0.0000
             13        C4H10      0.0000         0.0000

Binary Gas Diffusivity Option:  Wilke-Lee Modification to Chapman-
                                Enskog
Binary Liquid Diffusivity Option:  Wilke-Chang Modification to
                                   Stokes-Einstein EON
Mixture Liquid Diffusivity Option:  Modification of Holmes et al.
                                    to Wilke-Chang Equation
Transfer Coefficient Option:  ONDA et al.

  Packing Height          7.10 Feet
  Packing Type            1/4 Inch Ceramic Intalox Saddles
  Packing Increm          10.0
  Int. Area Corr.         1.00
  Maximum Iterations      9
  Convergence Toleranc    0.0000100  0.0001000  0.0150000  0.0050000
  Convergence Accelera    0.50000  0.50000
```

Table 11 (continued)

```
z(ft)  comp  y(bulk)  x(bulk)  x(inter) y(inter)

0.0000 CO2   0.22134  0.02997  0.05487  0.18632
       H2S   0.00170  0.00023  0.00082  0.00065
       COS   0.00010  0.00001  0.00004  0.00005
       MeOH  0.00000  0.96604  0.93860  0.00109  DYDZ(KEY)= -4.17226
       H2    0.32927  0.00071  0.00082  0.34498  DTGDZ =  -306.67328
       CO    0.17243  0.00074  0.00135  0.17720  FGKEY*AA = 92.75710
       N2    0.21124  0.00061  0.00106  0.22295  FLKEY*AA =159.79678
       CH4   0.00450  0.00047  0.00061  0.00448  HLAA =  21530.7
       C2H6  0.00650  0.00064  0.00081  0.00662  TL =         9.58
       C3H8  0.00000  0.00000  0.00000  0.00000  TG =        42.53
       TOT   1.00000  1.00000  1.00005  0.99948

0.0948 CO2   0.20413  0.02706  0.05246  0.16723
       H2S   0.00121  0.00016  0.00059  0.00044
       COS   0.00008  0.00001  0.00003  0.00004
       MEOH  0.00046  0.96926  0.94107  0.00092  DYDZ(KEY) = -4.14992
       H2    0.33722  0.00069  0.00083  0.35410  DTGDZ =     -51.34142
       CO    0.17611  0.00067  0.00139  0.18116  FGKEY*AA =  89.38692
       N2    0.21601  0.00055  0.00109  0.22852  FLKEY*AA = 156.15413
       CH4   0.05378  0.00052  0.00110  0.05606  HGAA =     1314.79565
       C2H4  0.00449  0.00045  0.00063  0.00442  HLAA =    21246.5
       C2H6  0.00651  0.00062  0.00083  0.00641  TL =           6.93
       C3H6  0.00000  0.00000  0.00000  0.00000
       C3H8  0.00000  0.00000  0.00000  0.00000
       C4H1  0.00000  0.00000  0.00000  0.00000  TG =          13.47
       TOT   1.00000  1.00000  1.00003  0.99930

0.1900 CO2   0.18613  0.02413  0.04827  0.15005
       H2S   0.00085  0.00011  0.00042  0.00030
       COS   0.00006  0.00001  0.00003  0.00003
       MEOH  0.00065  0.97251  0.94537  0.00086  DYDZ(KEY) = -3.87817
       H2    0.34569  0.00067  0.00085  0.36245  DTGDZ =     -18.17164
       CO    0.17995  0.00059  0.00142  0.18486  FGKEY*AA =  87.15559
       N2    0.22105  0.00049  0.00111  0.23334  FLKEY*AA = 154.10341
       CH4   0.05465  0.00046  0.00112  0.05675  HGAA =     1300.21155
       C2H4  0.00446  0.00044  0.00063  0.00434  HLAA =    21090.8
       C2H6  0.00650  0.00060  0.00083  0.00622  TL =           5.56
       C3H8  0.00000  0.00000  0.00000  0.00000  TG =           8.58
       C4H1  0.00000  0.00000  0.00000  0.00000
       TOT   1.00000  1.00000  1.00004  0.99920
```

Table 11 (continued)

```
0.2920 CO2    0.16727  0.02119  0.04360  0.13278
       H2S    0.00057  0.00007  0.00028  0.00019
       COS    0.00005  0.00001  0.00002  0.00002
      MEOH    0.00074  0.97583  0.95014  0.00082   DYDZ(KEY) = -3.54506
        H2    0.35473  0.00064  0.00086  0.37102   DTGDZ =     -13.10440
        CO    0.18393  0.00048  0.00145  0.18857   FGKEY*AA =  85.03691
        N2    0.22636  0.00041  0.00113  0.23813   FLKEY*AA = 152.23471
       CH4    0.05548  0.00038  0.00114  0.05732   HGAA =     1287.99268
      C2H4    0.00440  0.00042  0.00063  0.00431   HLAA =    20949.9
      C2H6    0.00647  0.00058  0.00082  0.00603   TL =           4.35
      C3H6    0.00000  0.00000  0.00000  0.00000
      C3H8    0.00000  0.00000  0.00000  0.00000   TG =           6.73
      C4H1    0.00000  0.00000  0.00000  0.00000
       TOT    1.00000  1.00000  1.00008  0.99920

0.4035 CO2    0.14745  0.01822  0.03860  0.11516
       H2S    0.00036  0.00004  0.00018  0.00012
       COS    0.00003  0.00000  0.00002  0.00001
      MEOH    0.00077  0.97924  0.97854  0.00078   DYDZ(KEY) = -3.17118
        H2    0.36443  0.00061  0.00088  0.37980   DTGD7 =     -11.97280
        CO    0.18804  0.00035  0.00149  0.19225   FGKEY*AA =  82.87715
        N2    0.23195  0.00031  0.00116  0.24288   FLKEY*AA = 150.36354
       CH4    0.05624  0.00028  0.00116  0.05773   HGAA =     1275.81091
      C2H4    0.00430  0.00039  0.00062  0.00416   HLAA =    20808.7
      C2H6    0.00641  0.00055  0.00080  0.00580
      C3H6    0.00000  0.00000  0.00000  0.00000   TL    =        3.14
      C3H8    0.00000  0.00000  0.00000  0.00000   TG    =        5.26
      C4H1    0.00000  0.00000  0.00000  0.00000
       TOT    1.00000  1.00000  1.02343  0.99869

   PROGRAM LOOKING FOR MOL. FRACT. LESS THAN ZERO OR GREATER THAN 1.0
   OR IS IN A DISCONTINUOUS REGION

0.5282 CO2    0.12660  0.01524  0.03326  0.09715
       H2S    0.00021  0.00002  0.00010  0.00007
      MEOH    0.00002  0.00000  0.00001  0.00001
        H2    0.37490  0.98278  0.99999  0.00073   DYDZ(KEY) = -2.75716
        CO    0.19223  0.00017  0.00152  0.19625   FGKEY*AA = -10.70855
        N2    0.23784  0.00019  0.00118  0.24851   FLKEY*AA = 148.45987
       CH4    0.05689  0.00016  0.00117  0.05808   HGAA =     1263.23303
      C2H4    0.00417  0.00036  0.00060  0.00398   HLAA =    20665.2
      C2H6    0.00633  0.00052  0.00086  0.00613   TL =           1.92
      C3H6    0.00000  0.00000  0.00000  0.00000   TG =           3.77
      C3H8    0.00000  0.00000  0.00000  0.00000
      C4H1    0.00000  0.00000  0.00000  0.00000
       TOT    1.00000  1.00000  1.03960  1.00114
```

Table 11 (continued)

```
PROGRAM LOOKING FOR MOL. FRACT. LESS THAN ZERO (GREATER THAN 1.0
OR IS IN A DISCONTINUOUS REGION

0.6716 CO2   0.10463  0.01224  0.02751  0.07862
       H2S   0.00010  0.00001  0.00005  0.00003
       COS   0.00001  0.00000  0.00001  0.00001
      MEOH   0.00082  0.98653  0.99999  0.00069  DYDZ(KEY) = -2.29096
        H2   0.38643  0.00048  0.00092  0.39912  DTGDZ =      -9.53984
        CO   0.19649  0.00000  0.00152  0.19570  FGKEY*AA =  78.58609
        N2   0.24410  0.00002  0.00120  0.25273  FLKEY*AA = 146.48508
       CH4   0.05734  0.00000  0.00117  0.05774  HGAA =     1249.93164
      C2H4   0.00398  0.00032  0.00057  0.00373  HLAA =    20517.0
      C2H6   0.00609  0.00047  0.00082  0.00575  TL    =          0.69
      C3H8   0.00000  0.00000  0.00000  0.00000  TG    =          2.24
      C4H1   0.00000  0.00000  0.00000  0.00000
       TOT   1.00000  1.00006  1.03377  0.99442

PROGRAM LOOKING FOR MOL. FRACT. LESS THAN ZERO (GREATER THAN 1.0
OR IS IN A DISCONTINUOUS REGION

0.8442 CO2   0.08097  0.00921  0.02128  0.05955
       H2S   0.00004  0.00000  0.00002  0.00001
       COS   0.00001  0.00000  0.00000  0.00000
      MEOH   0.00085  0.99002  0.99999  0.00066  DYDZ(KEY) = -1.77516
        H2   0.39742  0.00037  0.00093  0.40754  DTGDZ  =     -5.71413
        CO   0.20268  0.00001  0.00152  0.19518  FGKEY*AA =  76.31802
        N2   0.24956  0.00000  0.00120  0.25268  FLKEY*AA = 144.63148
       CH4   0.05905  0.00000  0.00117  0.05743  HGAA  =    1236.50891
      C2H4   0.00370  0.00028  0.00053  0.00339  HLAA  =   20376.0
      C2H6   0.00572  0.00040  0.00077  0.00528  TL   =          -0.52
      C3H6   0.00000  0.00000  0.00000  0.00000  TG   =           0.59
      C4H1   0.00000  0.00000  0.00000  0.00000
       TOT   1.00000  1.00030  1.02741  0.98173

PROGRAM LOOKING FOR MOL.FRACT.LESS THAN ZERO (GREATER THAN 1.0
OR IS IN A DISCONTINUOUS REGION

1.0669 CO2   0.05573  0.00616  0.01465  0.04011
       H2S   0.00001  0.00000  0.00002  0.00001
       COS   0.00000  0.00000  0.00000  0.00000
      MEOH   0.00087  0.99339  0.99999  0.00062  DYDZ(KEY) = -1.22658
        H2   0.40792  0.00018  0.00094  0.41525  DTGDZ =      -7.12748
        CO   0.20916  0.00004  0.00152  0.19463  FGKEY*AA =  74.12832
        N2   0.25700  0.00000  0.00120  0.25263  FLKEY*AA = 142.81392
       CH4   0.06086  0.00000  0.00117  0.05709  HGAA  =    1221.90247
      C2H4   0.00329  0.00022  0.00047  0.00295  HLAA  =  20236.0
      C2H6   0.00516  0.00032  0.00069  0.00464  TL  =           -1.72
      C3H6   0.00000  0.00000  0.00000  0.00000  TG  =            0.59
      C3H8   0.00000  0.00000  0.00000  0.00000
      C4H1   0.00000  0.00000  0.00000  0.00000
```

Table 11 (continued)

```
        TOT    1.00000  1.00032  1.02065  0.96794

      PROGRAM LOOKING FOR MOL.FRACT.LESS THAN ZERO (GREATER THAN 1.0
      OR IS IN A DISCONTINUOUS REGION

 1.3892 CO2    0.02894  0.00309  0.00759  0.02031
        H2S    0.00001  0.00000  0.00002  0.00001
        COS    0.00000  0.00000  0.00002  0.00001
        MEOH   0.00092  0.99699  0.99197  0.00058  DYDZ(KEY) = -0.63822
        H2     0.41828  0.00000  0.00094  0.41736  DTGDZ =       2.86433
        CO     0.21635  0.00006  0.00152  0.19404  FGKEY*AA =  71.83870
        N2     0.26562  0.00000  0.00120  0.25256  FLKEY*AA = 140.88129
        CH4    0.06296  0.00001  0.00117  0.05674  HGAA  =   1203.14294
        C2H4   0.00265  0.00014  0.00037  0.00228  HLAA  =  20088.0
        C3H6   0.00000  0.00000  0.00000  0.00000  TL  =        -2.98
        C3H8   0.00000  0.00000  0.00000  0.00000  TG  =        -2.98
        C4H1   0.00000  0.00000  0.00000  0.00000
        TOT    1.00000  1.00050  1.00535  0.94757

 2.0087 CO2    0.00030  0.00000
        H2S    0.00002  0.00000
        COS    0.00000  0.00000
        MEOH   0.00000  1.00043
        H2     0.43254  0.00000
        CO     0.22373  0.00006
        N2     0.27468  0.00000
        CH4    0.06511  0.00001
        C2H4   0.00140  0.00000
        C2H6   0.00240  0.00000
        C3H6   0.00000  0.00000
        C3H8   0.00000  0.00000
        C4H1   0.00000  0.00000
        TOT    1.00047  1.00050

                           PROBLEM SOLVED IN 7 ITERATIONS
```

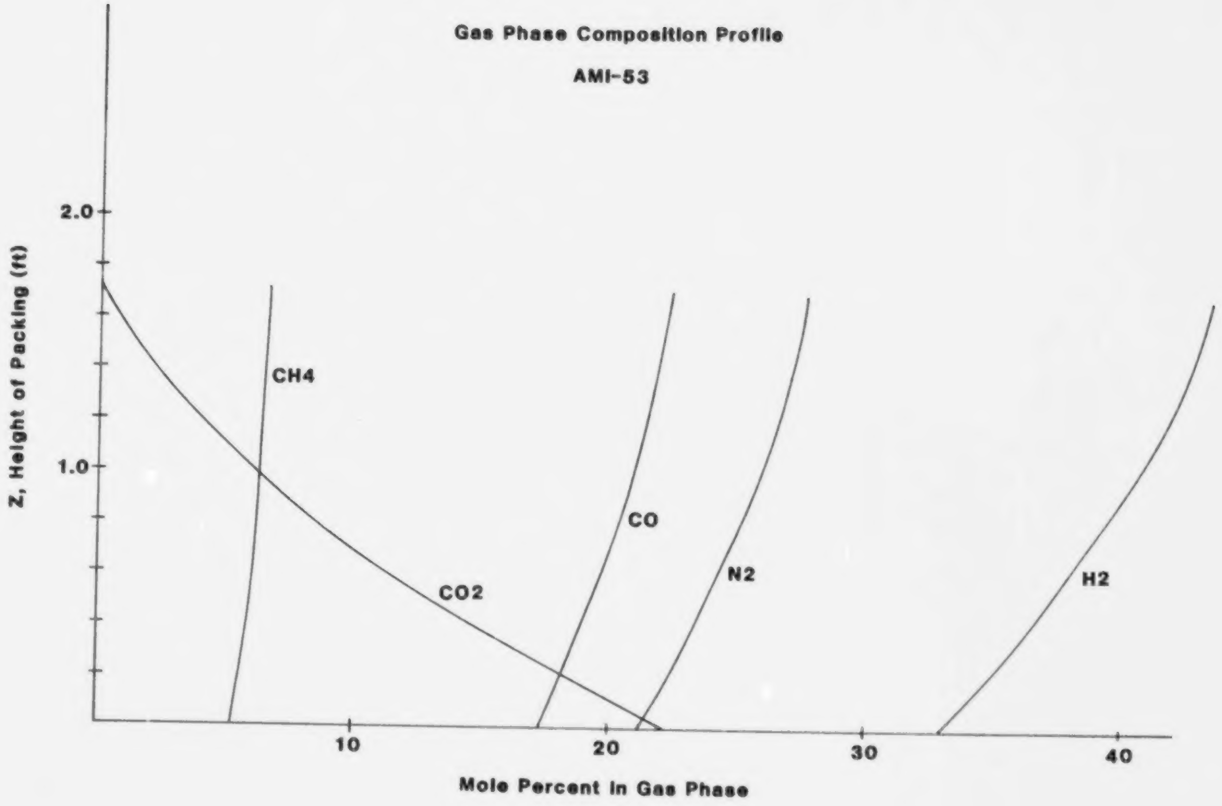
Ten packing increments were used. The gas temperature gradient, the molar flow gradient of the key component, (CO_2) as well as other pertinent information are given at each step of the calculation.

At each step, the composition of the gas phase at the interface is listed as is the total pressure which is calculated by summing the partial pressures of all components. The fact that the calculated total pressure differs from the system pressure is in part due to departure from ideal gas behavior. In addition, several of the gases are only slightly soluble in methanol at these conditions. The presence of very small amounts of these compounds at the liquid interface is associated with fairly high partial pressures of these compounds in the gas phase. For instance, a liquid phase concentration of 1000 parts per million (volume) of H_2 will be in equilibrium with a gas phase partial pressure of almost 8 atmospheres at 0°F. The sensitivity of the calculated total pressure to the quantity of these gases dissolved in the liquid is in part responsible for the difference between the calculated and actual pressure as well as the failure of the interfacial gas mole fractions to sum to 1.0.

Figures 54 and 55 show the variation in composition of the gases as they move through the absorber for AMI-53. Two graphs are presented because of the range of compositions encountered; note that the acid gases, H_2S, COS, and CO_2, are readily absorbed while the product gases, CO, H_2, and CH_4, increase in concentration through the column, mainly due to the concentrating effect of removing large amounts of the acid gases.

It is interesting to note that the specified recovery of CO_2, the key component, was achieved at the 1.7 foot level of packing. At this point, most of the gas phase concentrations were changing little with packing height. Ethane and ethylene, however, were still transferring at a fairly rapid rate as evidenced by the shape of their concentration profiles in Figure 55. At the bottom of the column, these two gases were being absorbed but the concentration profiles also reflect the concentrating effect of removing large amounts of CO_2. Moving up the column, the composition profiles for ethane and ethylene become steeper, especially after most of the CO_2 is removed, and indicate that their absorption was still taking place at the point where the calculation was terminated. In fact, while the computer prediction shows that approximately 71.1% of the ethylene and 67.2% of the ethane was absorbed, experimental data show that 96.5% of the ethylene and 94.7% of the ethane were actually recovered. Clearly, the choice of ethane or ethylene as the key component would provide a better picture of what is happening in the entire 7.1 feet of packing used in this experimental run.

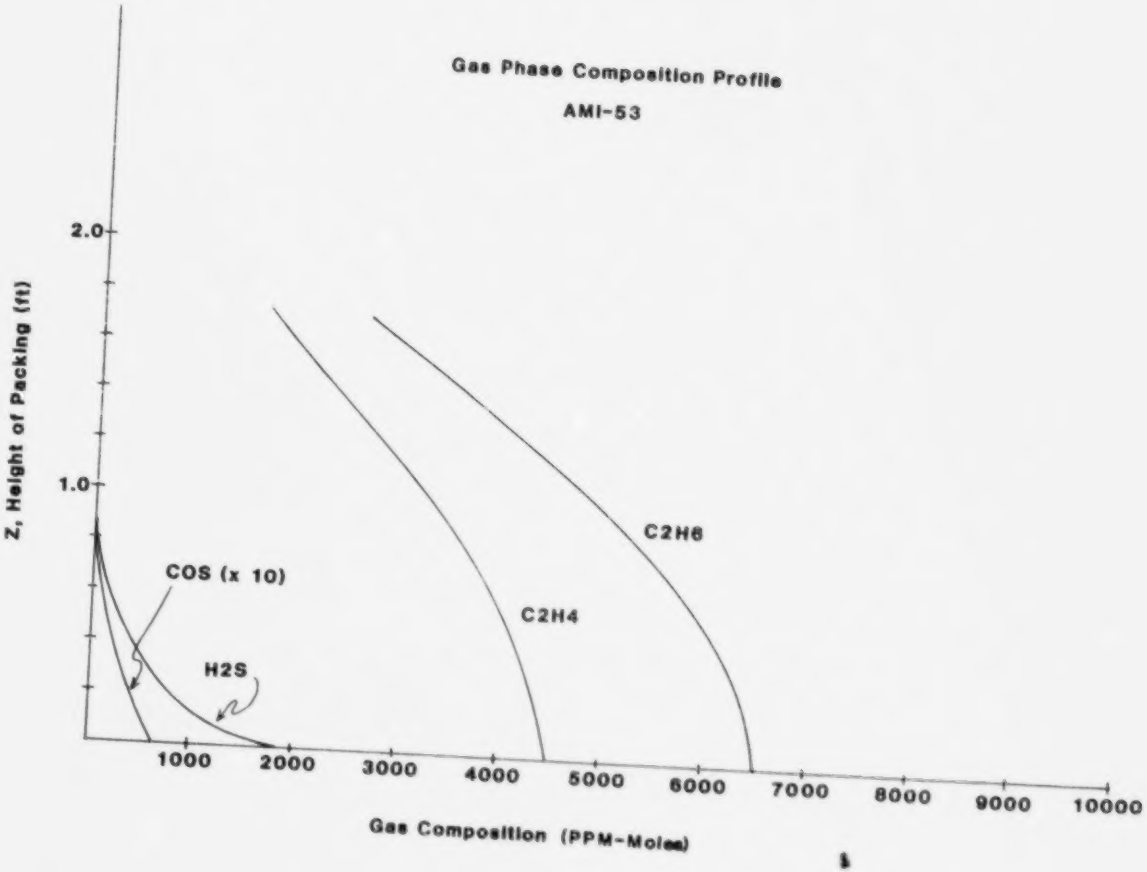
Figure 54



Gas Phase Composition Profile

AMI-53

Figure 55


Gas Phase Composition Profile
AMI-53
Z, Height of Packing (ft)
2.0
1.0
COS (x 10)
H2S
C2H4
C2H6
1000
2000
3000
4000
5000
6000
7000
8000
9000
10000
Gas Composition (PPM-Moles)

To explore this aspect more fully, ethane was chosen as the key component and the computer model run with ethane recovery set at 94.7%. Convergence problems prevented any useful information from being obtained. These convergence problems stemmed from the fact that for the specified recovery of ethane, the L/G ratio is close to or below the minimum if ideal gas behavior is assumed. By reducing the specified recovery of ethane to 67.2%, model convergence resulted.

The effect of column conditions on the product of the mass transfer coefficient and the interfacial area is shown in Figure 56, which also shows the variation of this quantity with packing height for both liquid and gas phases for the key component, CO_2. It is interesting to note that while the product of gas-phase mass transfer coefficient and interfacial area ranged from 93 to 69 lb-moles/hr-ft^2, the height of a gas-phase transfer unit, HTU_G varied only from 0.191 to 0.195 feet. The product of the liquid-phase mass transfer coefficient and the interfacial area ranged from 161 to 143 lb-moles/hr while the height of a liquid-phase transfer unit varied only from 0.823 to 0.900 feet. This result confirms accepted design approximations that assume heights of transfer units to be relatively constant through a column. Approximately the same degree of variation was noted for the product of the individual heat transfer coefficients and the interfacial area, as is shown in Figure 57. This similarity is not surprising since the j-factor analogy was used to estimate these transport coefficients.

While it appears that the heights of transfer units for CO_2 absorption in AMI-53 are relatively constant, the number of transfer units will change through the column in response to the changing temperature and phase equilibrium relationships. This is illustrated in Figure 58 which shows a McCabe-Thiele diagram for both the isothermal and adiabatic operation. The significance of these thermal effects was evaluated by running the model assuming the heat of solution of all components in methanol to be zero and by assuming that the gas enters at the same temperature as the entering solvent. The calculated height for the isothermal case was 1.57 feet as compared to 2.01 feet required for the adiabatic case; thus, isothermal operation will require only 79% of the packed height required for the adiabatic case. The importance of the thermal effects becomes more significant as the minimum L/G ratio is approached. ieces of information that can be gained from the use of the model. Additionally, if analysis rather than a design approach is needed, the removal efficiency of the key component can be increased or decreased so that, for instance, 7.1 feet is the calculated height of packing.

Figure 56

AMI-53
ONDA Correlation

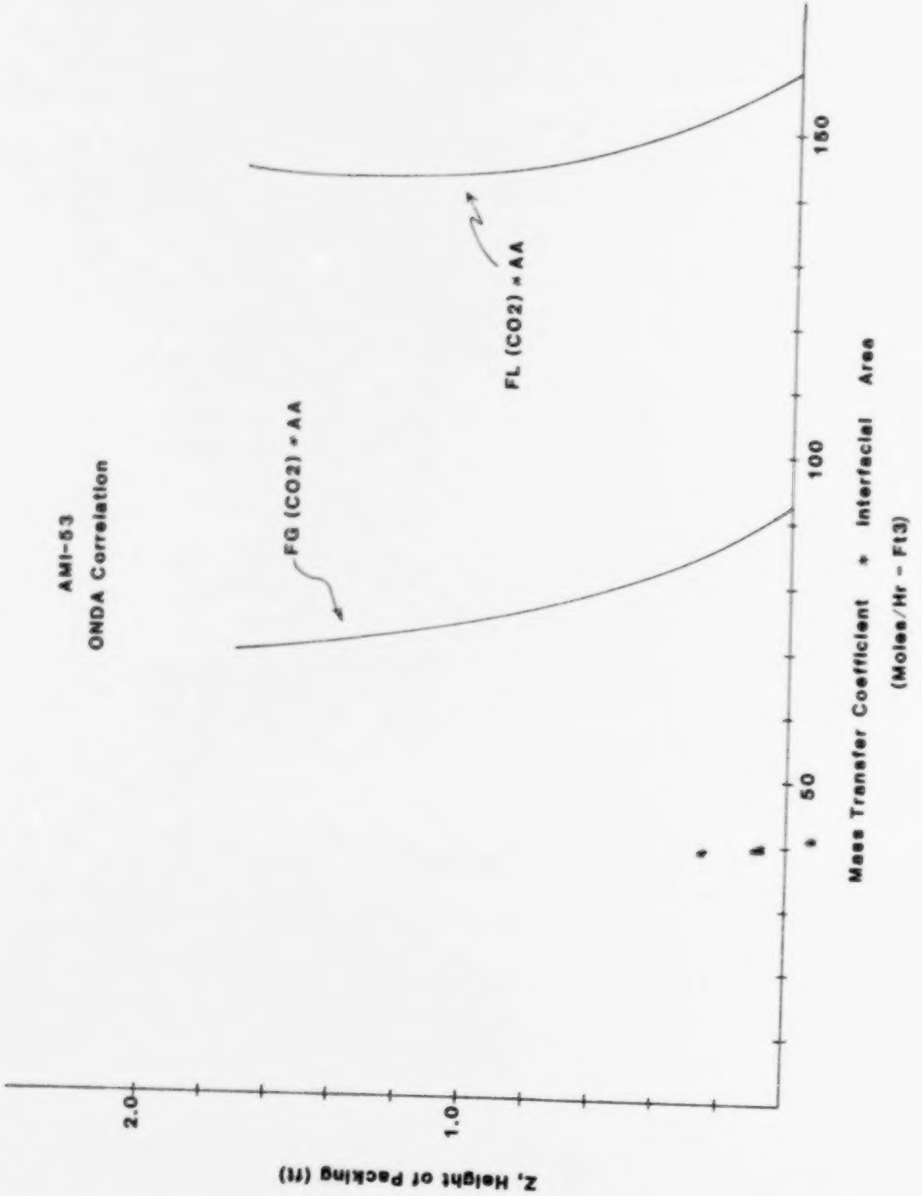


Figure 57

AMI-53

ONDA Correlation

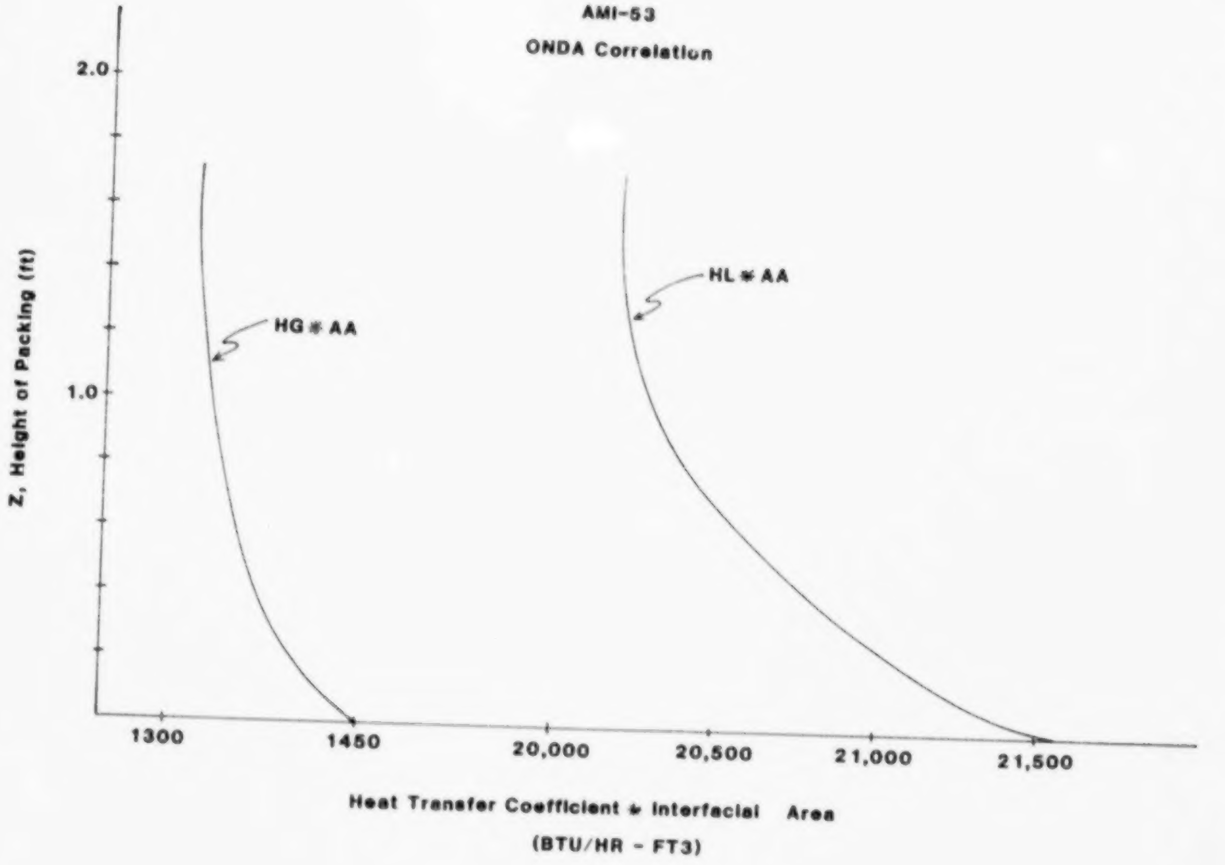
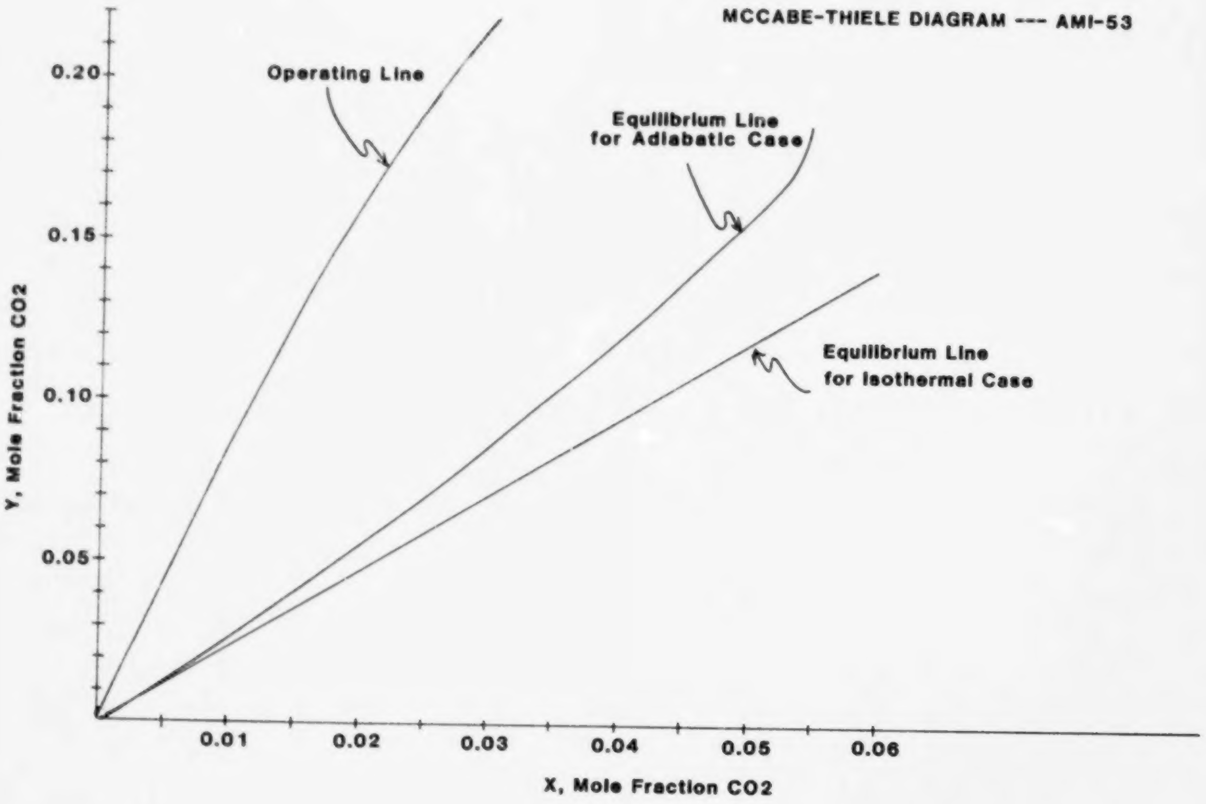


Figure 58


MCCABE-THIELE DIAGRAM --- AMI-53
Operating Line
Equilibrium Line
for Adiabatic Case
Equilibrium Line
for Isothermal Case
0.20
0.15
0.10
0.05
Y, Mole Fraction CO2
0.01
0.02
0.03
0.04
0.05
0.06
X, Mole Fraction CO2

MINIMUM LIQUID-TO-GAS RATIO

Specification of the liquid flow rate is required for absorber design. The minimum liquid-to-gas ratio may be evaluated from physical property data and it provides a useful limit in setting the design flow rates. For a given inlet gas flow rate and composition, the minimum liquid rate can be defined as the lowest liquid rate that will allow the specified recovery of a given solute without the formation of a "pinch point" in the column. A "pinch point" occurs when the driving force for mass transfer of the solute from the gas phase to the interface becomes zero; that is, if mole fractions are used in the driving force expression, the mole fraction of the solute at the gas-phase interface is equal to the mole fraction of the solute in the bulk gas phase.

Feintuch (1973) modified his computer algorithm to estimate the minimum liquid rate required for a given inlet gas flow rate and composition, a specified solute recovery, and a given set of column operating conditions. His approach was to find the minimum liquid flow rate that allowed the calculation to reach the top of the tower with the driving force of the solute to the interface, remaining positive at all times. Because the algorithm used here and by Feintuch will not converge at or below the minimum liquid rate for the key component, it is important to know this quantity. The algorithm will, however, converge for multicomponent cases where the liquid flow is at or below the minimum for non-key component recovery.

Adopting the method of Feintuch, SIMPAK was modified to estimate the minimum liquid rate for all Syngas and Integrated runs made for this study. The subroutine MINLG, written by Feintuch (1978) was included in the computer program SIMPAK. This subroutine systematically brackets the minimum liquid rate until it can be determined to within 0.5 lbmoles/hr/ft^2. The modified version of SIMPAK, similar to the one used by Feintuch, is given by Kelly (1981).

In the comparison of experimental data to predicted results for both Syngas and Integrated runs, four cases would not converge. It was thought at the time that these runs were probably at or below the minimum liquid- to-gas ratio. The data from all these runs were entered into the program, SIMLG, to estimate this ratio. In the case of the Integrated runs, CO_2 was designated as the key component and the remainder of the gases were grouped together and entered as N_2. Although the minimum L/G algorithm could be implemented for the multicomponent case, excessive computation times would be required. The results from these computer runs can be found in Tables 12 and 13.

It is interesting to note that for three of the four runs that failed to converge, AM-27, AMI-44, and AM-55, operation was at or below the minimum liquid rate. Run AMI-57 was sufficiently above the minimum but failed to converge for no apparent reason. It is possible that optimization of the choice of convergence criteria could yield better results. Also, it is interesting to note that model convergence occurred for cases ranging from 1.19 to 4.24 times the minimum liquid rate. The choice of CO_2 as the key component was made for the same reasons as before; namely, its large concentration and high solubility make it easier to monitor its recovery in the column. In the future, incorporating this technique into MCOMP would make possible the estimate of the minimum liquid rate required for the recovery of other key components in the presence of large heat effects accompanying CO_2 absorption. Because of the large amounts of time required for model convergence for multicomponent cases, it was decided not to pursue this objective in the study. Modification of MCOMP for this purpose is, however, straightforward.

It appears that model convergence for cases approaching the minimum liquid-to-gas ratio is possible although the closer the limit is approached, the more difficult convergence becomes. With some refinement of the current convergence criteria and the prudent use of contingencies at points in the computation where the driving force may reverse, convergence close to the minimum L/G could be accomplished. For cases below the minimum liquid rate, convergence is still possible if another key component is chosen and its removal efficiency set at a point where the recovery of the component of interest was. This would allow for the prediction of temperature maxima in the column, which is a case closely studied by Stockar and Wilke (1977). This also allows for cases where stripping of non-key components is taking place arising from thermal effects.

TABLE 12

MINIMUM L/G RATIOS

RUN	L	T_L	G	T_G	P	CO_2 RECOVERY	$\frac{L/G}{\{L/G\}_{min}}$
24	69.39	-33.69	17.79	48.83	450	0.999	1.89
25	70.43	-34.02	18.16	45.56	300	0.993	1.45
26	57.48	-33.30	17.30	46.58	300	0.997	1.19
27	50.82	-28.55	17.30	52.46	300	0.998	1.00*
32	61.18	-31.79	17.31	57.42	400	0.981	1.50
33	72.05	-28.91	16.92	53.18	400	0.983	1.78
34	54.38	-29.33	16.99	61.24	400	0.975	1.38
38	91.77	-18.62	17.11	56.48	400	0.998	2.10
50	127.56	- 6.33	17.50	40.23	250	0.998	1.93
55	47.91	- 4.17	17.83	57.07	250	0.701	0.94*
30	60.82	-28.27	16.21	54.04	400	0.958	1.71
35	60.82	-34.65	15.86	53.86	400	0.991	1.72
36	72.76	-27.75	16.39	57.50	300	0.984	2.06
37	71.42	-22.20	16.64	59.92	450	0.977	1.80
43	63.55	-34.07	18.08	40.13	250	0.990	1.30
44	63.51	0.84	16.96	47.62	250	0.950	1.04*
45	121.32	-34.07	16.99	22.31	250	0.990	4.24
47	130.48	-33.07	17.11	36.33	450	0.990	3.93
52	127.87	-30.70	16.83	38.05	450	0.999	3.92
53	127.46	- 4.28	17.88	42.53	250	0.990	1.99
57	61.66	-32.70	15.39	50.44	450	0.990	2.03*
59	127.30	- 2.92	14.91	44.23	250	0.986	2.36
60	100.80	-16.00	16.04	56.39	350	0.999	2.42

* NO CONVERGENCE

L,G	LB-MOLES/HR/FT^2
T_L,T_G	DEGREES F
P	PSIG

HEIGHT OF PACKING IN FEET

TABLE 13

MINIMUM L/G RATIOS

RUN	L	L(MIN)	G	L/G	L/G (MIN)	$\frac{L/G}{(L/G)_{min}}$
24	69.39	36.64	17.79	3.90	2.06	1.89
25	70.43	48.60	18.16	3.88	2.68	1.45
26	57.48	48.50	18.01	3.19	2.69	1.19
27	50.82	50.80	17.30	2.94	2.94	1.00*
32	61.18	40.70	17.31	3.53	2.35	1.50
33	72.05	40.50	16.92	4.26	2.39	1.78
34	54.38	39.30	16.99	3.20	2.31	1.38
38	91.77	43.80	17.11	5.36	2.55	2.10
50	127.56	66.00	17.50	7.29	3.77	1.93
55	47.91	50.90	17.83	2.69	2.85	0.94*
30	60.82	35.50	16.21	3.75	2.19	1.71
35	60.82	35.40	15.86	3.83	2.23	1.72
36	72.76	35.38	16.39	4.44	2.16	2.06
37	71.42	39.70	16.64	4.29	2.39	1.80
43	63.55	48.91	18.08	3.51	2.71	1.30
44	63.51	61.13	16.96	3.74	3.60	1.04*
45	129.32	30.50	16.99	7.61	1.80	4.24
47	130.48	32.90	17.11	7.63	1.94	3.93
52	127.87	32.70	16.83	7.60	1.94	3.92
53	127.46	64.04	17.88	7.13	3.58	1.99
57	61.66	30.42	15.39	4.01	1.98	2.03*
59	127.30	53.98	14.91	8.54	3.62	2.36
60	100.80	41.60	16.04	6.28	2.59	2.42

* NO CONVERGENCE

L,G LB-MOLES/HR/FT^2

COMPARISON OF TRANSFER COEFFICIENT CORRELATIONS

The correlations of Onda et al (1968) were used to estimate mass transfer coefficients and interfacial area in the computer runs comparing model predictions to experimental results. This choice was based on the fact that this set of correlations had been developed with data obtained for small packing sizes and for cases where organic liquids, including methanol, were used. Results presented earlier showed that this set of correlations yielded excellent agreement between predicted and experimental data.

In order to evaluate the accuracy of using other sets of correlations found in the literature, four options were programmed into the model to be chosen in the program input:

OPTION 1	SHULMAN
OPTION 2	ONDA
OPTION 3	FAIR
OPTION 4	SHULMAN/PURANIK AND VOGELPOHL

To illustrate the use of these options, the computed liquid temperature profile was compared to the results from syngas runs AM-32 and AM-50. These results are shown graphically in Figures 59 and 60, and are tabulated in Table 14 where the range of values over the entire packing height are reported.

Several points can be made in examining the results obtained from the four transfer coefficient-interfacial area correlations. The comparison of the four options was made on the basis of the product of the transfer coefficient and the interfacial area, which should be easier to correlate than either quantity taken separately. In this respect, the approach taken by Fair (1981) seems to be the most sound as it correlates the product of the transfer coefficient at the interfacial area. However, Bolles and Fair (1981) suggest that the scatter in the data used to develop this correlation requires the use of a safety factor of 1.70. In the same study they also suggest using a safety factor of 2.23 with the Onda correlation.

Figure 59

AM-32
Comparison of Transfer Coefficient Options

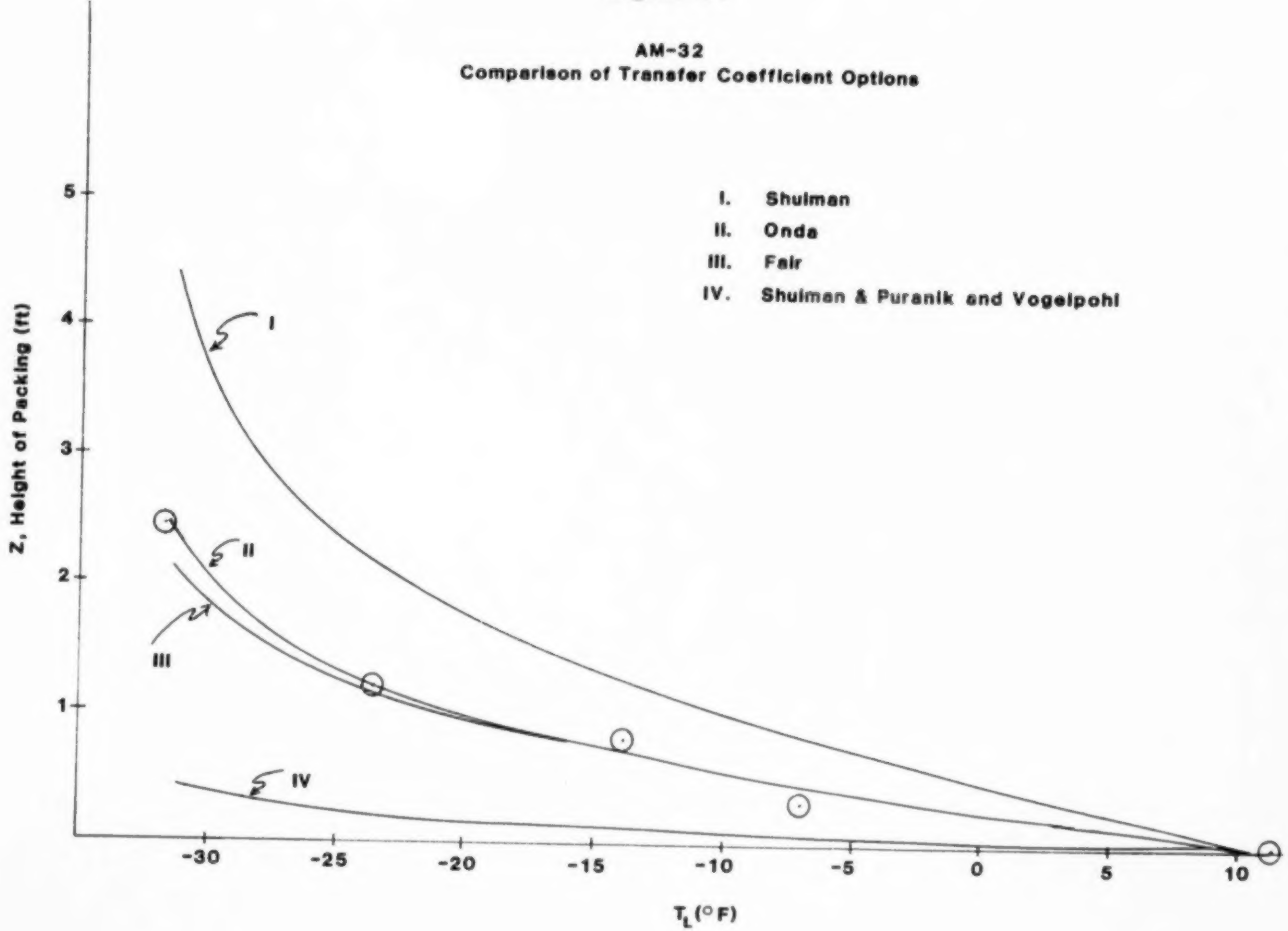


Figure 60

AM-50

Comparison of Transfer Coefficient Options

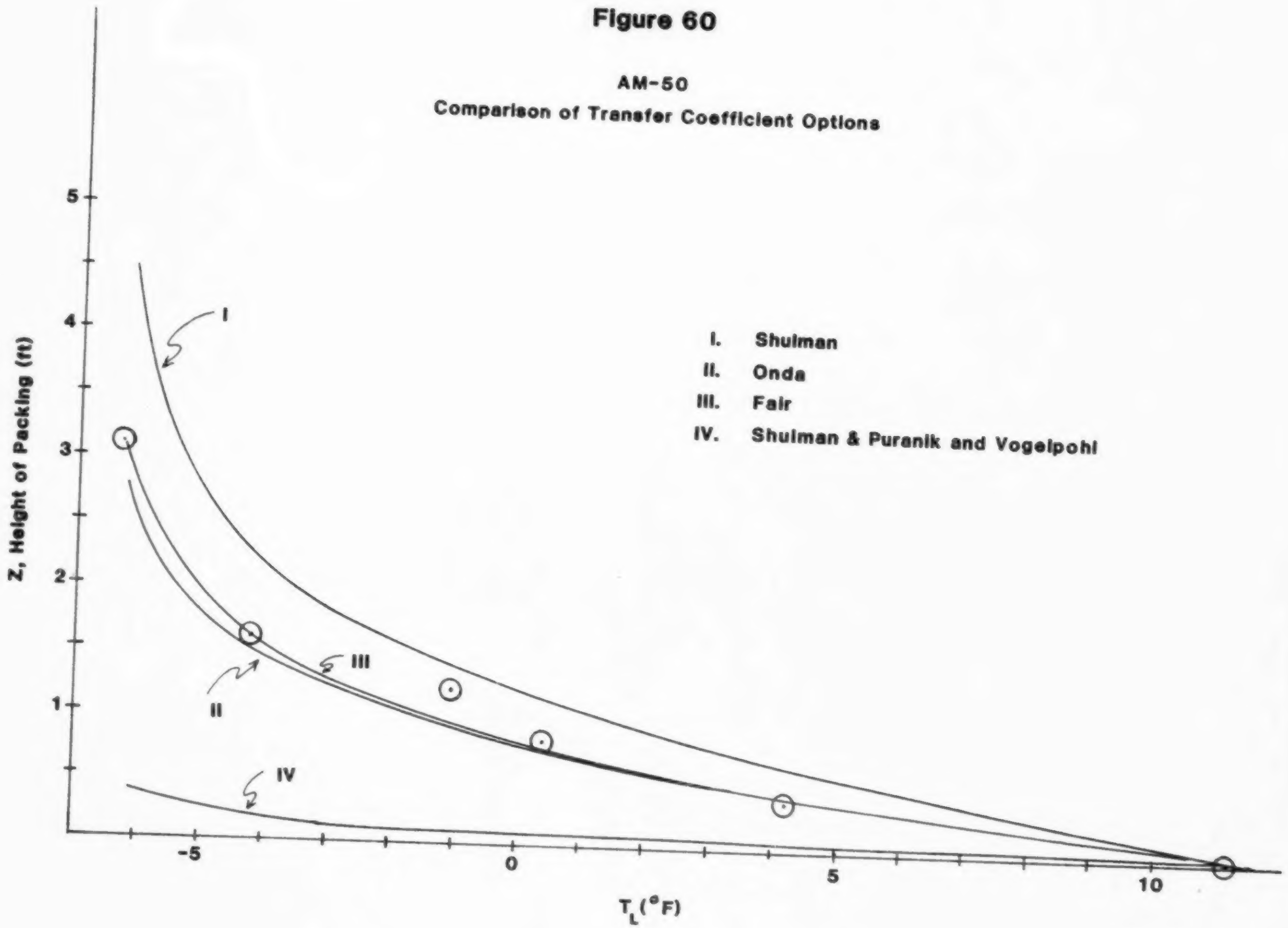


TABLE 14
TRANSFER COEFFICIENT OPTIONS --- RESULTS

	OPTIONS
1	SHULMAN
2	ONDA
3	FAIR
4	SHULMAN/PURANIK AND VOGELPOHL

RESULTS FROM AM-50

OPTION	FG*AA	FL*AA	HG*AA	HL*AA	HEIGHT
1	28-19	196-166	220-159	26191-25318	4.63
2	72-47	164-130	561-392	21869-19818	2.90
3	129-77	136-78	1000-616	18007-11810	3.21
4	317-210	2202-1834	2470-17516	294036-279494	0.42

RESULTS FROM AM-32

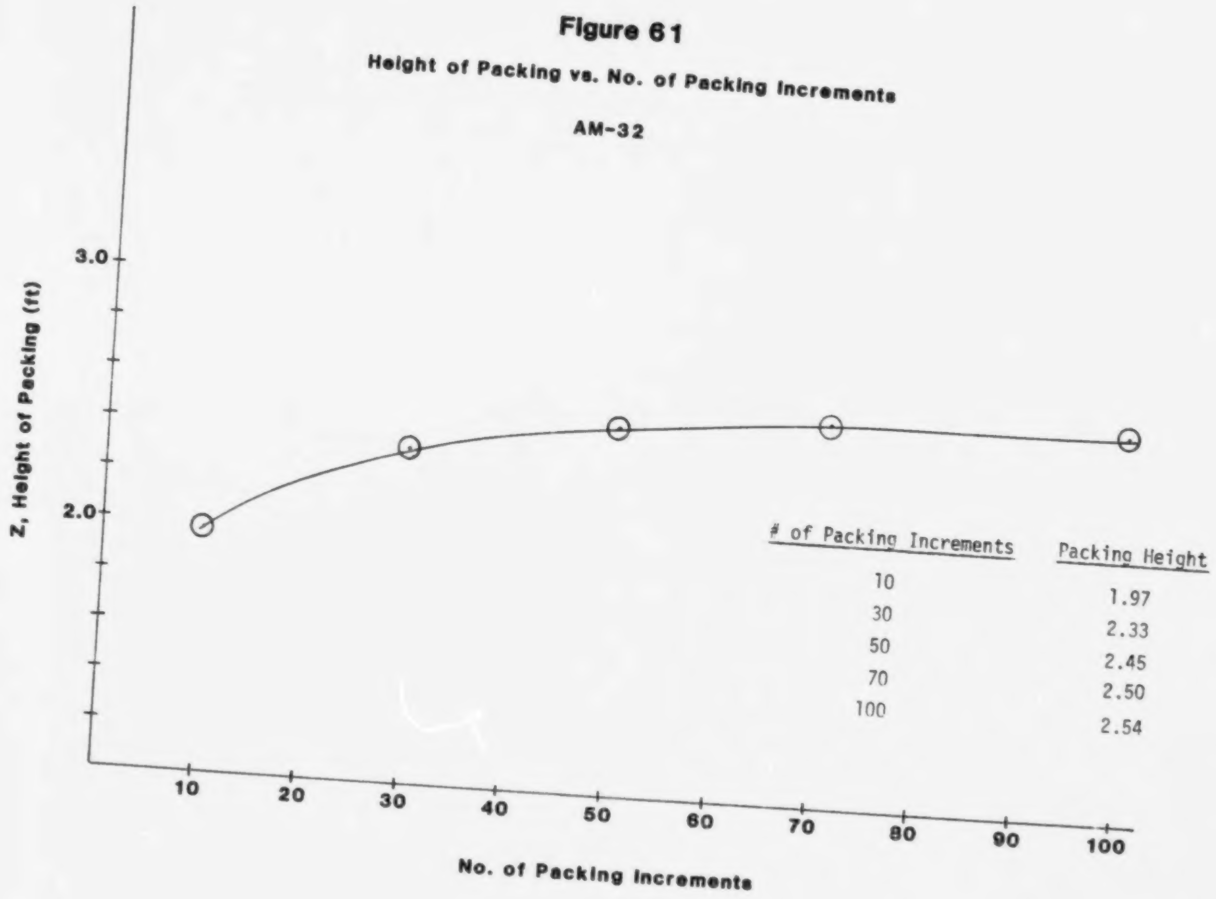
OPTION	FG*AA	FL*AA	HG*AA	HL*AA	HEIGHT
1	21-14	109-78	165-117	14364-13844	4.56
2	62-38	100-59	491-320	13143-10458	2.45
3	161-101	66-41	1269-814	8547-7287	2.22
4	229-139	1202-787	1819-1184	158664-139516	0.44

FG*AA, FL*AA	LB-MOLES/HR/FT^3
HG*AA, HL*AA	BTU/HR/FT^3
HEIGHT	FEET

Figure 61

Height of Packing vs. No. of Packing Increments

AM-32



# of Packing Increments	Packing Height
10	1.97
30	2.33
50	2.45
70	2.50
100	2.54

The other three options represent separate correlations of effective interfacial area and transfer coefficient. Table 15 shows the wide discrepency between the values of the mass transfer coefficient and the interfacial area for options 1, 2 and 4. These differences underline the variability in efforts to correlate these two quantities separately. As is evident from Figures 59 and 60, combining the interfacial area correlation of Puranik and Vogelpohl (1974) with the mass transfer coefficient correlation of Shulman (1955), resulted in poor representations of the measured temperature profile.

From the results obtained in this study, it appears there are significant differences between the results of various researchers in developing generalized correlations for mass transfer in packed columns. This is partly the result of the scatter in the data used to develop these correlations. While the approach used by Fair avoids the problem of correlating the interfacial area and transfer coefficient separately, an effective generalized correlation for each quantity would be more desirable so that extension of these results can be made to a wider variety of conditions. At this point, it appears that additional experimental data and better methods of correlation are needed to improve the design procedures for packed columns.

TABLE 15
TRANSFER COEFFICIENT OPTIONS --- RESULTS

	OPTIONS
1	SHULMAN
2	ONDA
3	FAIR
4	SHULMAN/PURANIK AND VOGELPOHL

RESULTS FROM AM-50

OPTION	FG	FL	AA	AA/AA(TOTAL)
1	2.34-1.64	16.20-14.31	12.1-11.6	0.04
2	0.34-0.23	0.78-0.64	210.7-202.6	0.69
3				
4	2.34-1.64	16.20-14.31	135.7-128.3	0.44

RESULTS FROM AM-32

OPTION	FG	FL	AA	AA/AA(TOTAL)
1	2.27-1.57	11.76-8.75	9.3-8.9	0.03
2	0.34-0.23	0.55-0.36	180.9-164.1	0.58
3				
4	2.27-1.57	11.76-8.75	102.4-89.8	0.32

FG, FL	LB-MOLES/HR/FT^2
AA, AA(TOTAL)	FT^2/FT^3

RECOMMENDATIONS FOR APPLICATION OF MODEL

The mathematical model of the packed absorber described here has been used to analyze the results of the experimental runs made for the purpose of this study. Other objectives in developing this model included use of the model to extend the experimental results to situations not attainable in pilot plant operation. Future work will extend the packed absorber model to stripper operation and culminate in the development of a simulation package to describe the performance of the entire system.

At this point, it would be useful to illustrate several ways in which the packed absorber model could be used in the perspective of acid gas removal. Two examples of the application of the model to situations encountered in acid gas removal will be presented here for this purpose.

PARAMETRIC STUDIES

One of the more valuable uses of a computer model is to evaluate the effect of changes in process variables on the final column design. This may require a complex optimization procedure or may be fairly simple if only one variable is involved. In either case, the computer model becomes the tool for the evaluation.

As a simple example of how the models developed here might be used for a parametric study, the operating conditions for Syngas runs AM-32 and AM-50 were used as a base case for the computer model SIMPAK. The inlet liquid temperature, inlet gas temperature, inlet liquid flow rate, and the operating pressure were all varied to see how this would effect the calculated packing height.

Figure 62 shows the effect of the inlet liquid temperature on the height of packing for runs AM-32 and AM-50. All other conditions were the same as those used in the experimental runs. Over the ranges shown here, a change from -60^{o}F to 18^{o}F for AM-50 resulted in only a 0.3 foot change in the required packing height. This was due to the high liquid rate used in this run. For AM-32, in which a much lower liquid rate was used, the effect of changing the inlet liquid temperature was more pronounced, especially at temperatures greater than 0^{o}F. For a liquid temperature change from 0^{o}F to 20^{o}F, the required height of packing changes about 1.90 feet. Knowledge of the sensitivity of the design estimate to process changes can be important in the development of an economically feasible process.

Figure 62

Packing Height vs. Lean Solvent Temperature

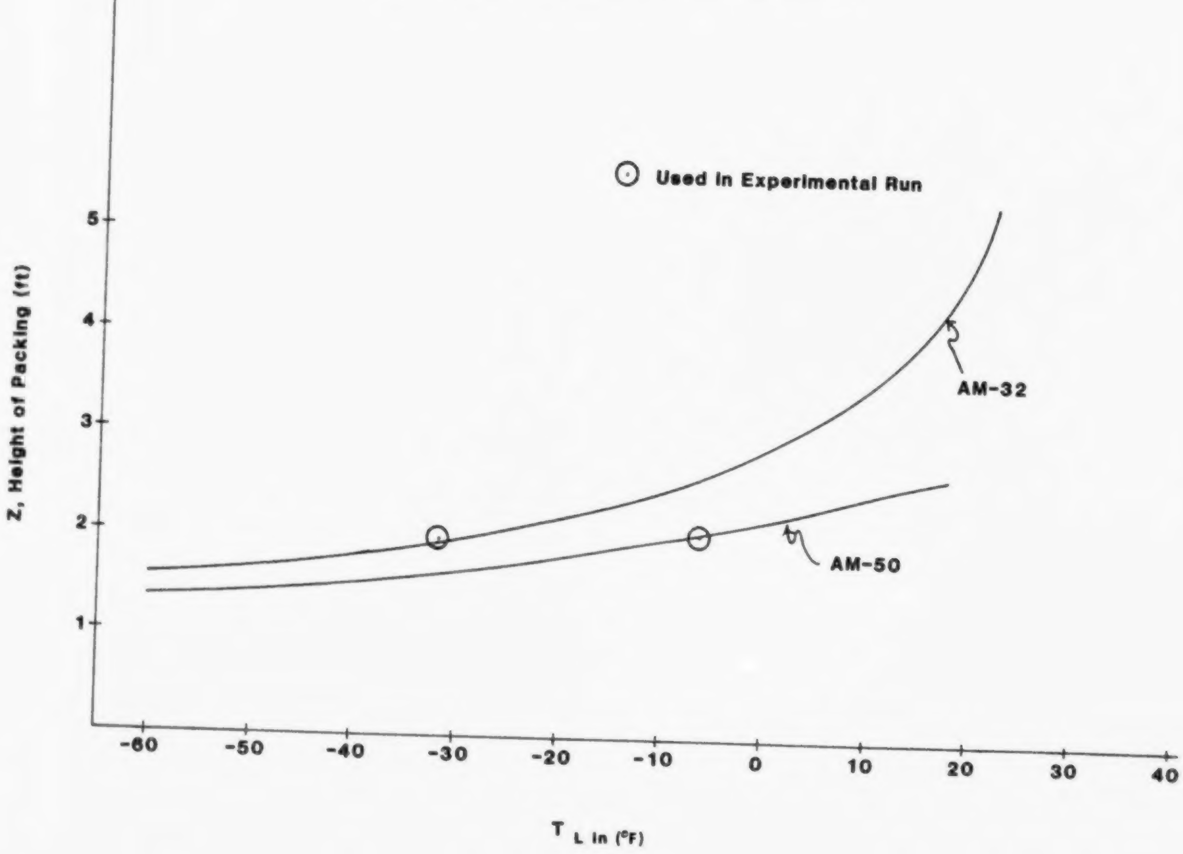


Figure 63 shows the effect of inlet gas temperature on the height of packing for runs AM-32 and AM-50. In both cases, as the gas temperature changes from 10°F to 140°F, the required packing height changes less than 0.1 foot. The relatively small amount of sensible heat associated with the inlet gas stream explains the insensitivity of packing height to changes in this parameter.

The effect of liquid flow rate on packing height is shown in Figure 64. The fact that AM-50 was made at a higher inlet liquid temperature is partly responsible for the greater sensitivity of the packing height to the liquid flow rate. The solubility of CO_2 in methanol at the inlet liquid temperature of AM-50 is much less than is the case for AM-32.

Figure 65 shows the sensitivity of the packing height to changes in system pressure. System pressure has a large effect on the solubility of CO_2 in methanol which is evident from the information in Figure 64. For AM-50, as the pressure is reduced from 18 to 11 atmospheres, the packing height requirements change from 2.0 to 3.9 feet while a change from 50 to 20 atmospheres requires only an increase of 0.7 feet. This reflects the reduced capacity of the solvent for CO_2 at the lower pressures and the increased difficulty in recovering the specified amount of solute.

While these four examples are relatively simple, they nonetheless demonstrate the value of a computer model for process analysis and design. This type of model can also be useful in developing process control strategy for non-linear systems.

EFFECT OF STRIPPING OPERATION ON ABSORBER

In any process where an absorber and stripper are used to recover a solute and then regenerate the solvent, one operation can significantly affect the other. This is especially true in situations where the stripper does not effectively regenerate the solvent that is fed to the absorber. The presence of any component that is to be recovered in the solvent entering the absorber reduces the driving force for mass transfer and hence lowers the recovery of that component.

For the runs made in this study, computer predictions indicated that if clean methanol were fed to the absorber, the recovery of the H_2S in the entering gas stream would be nearly complete. Experimental results, however, showed significant concentration of H_2S in the absorber exit gas. While qualitative analysis did show that there was some H_2S in the stripped methanol entering the absorber, sampling problems prevented the determination of the actual concentration.

To investigate this problem, the computer model MCOMP was used to analyze several hypothetical cases in which significant amounts of H_2S were present in the methanol fed to the absorber. The conditions used in the program input were the same as in AMI-59 with the exception that the H_2S

Figure 63

Packing Height vs. Sour Gas Inlet Temperature

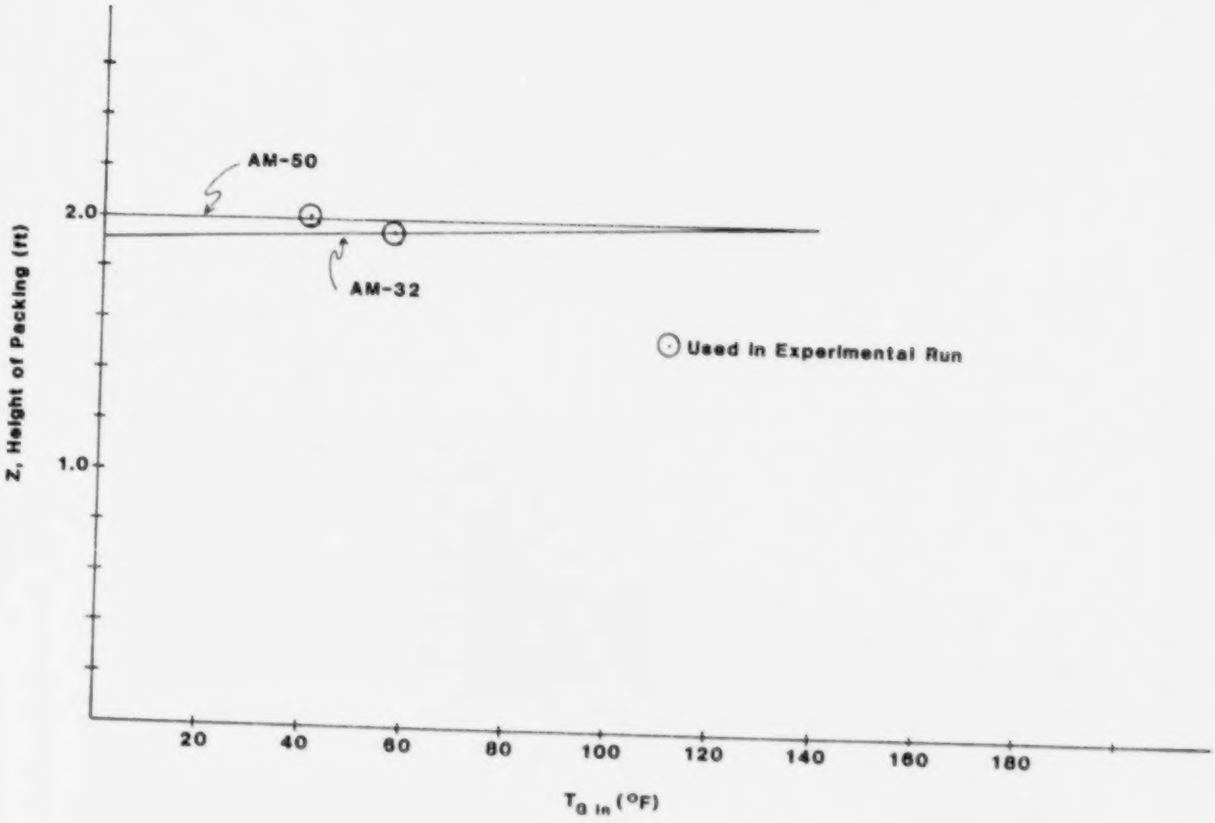
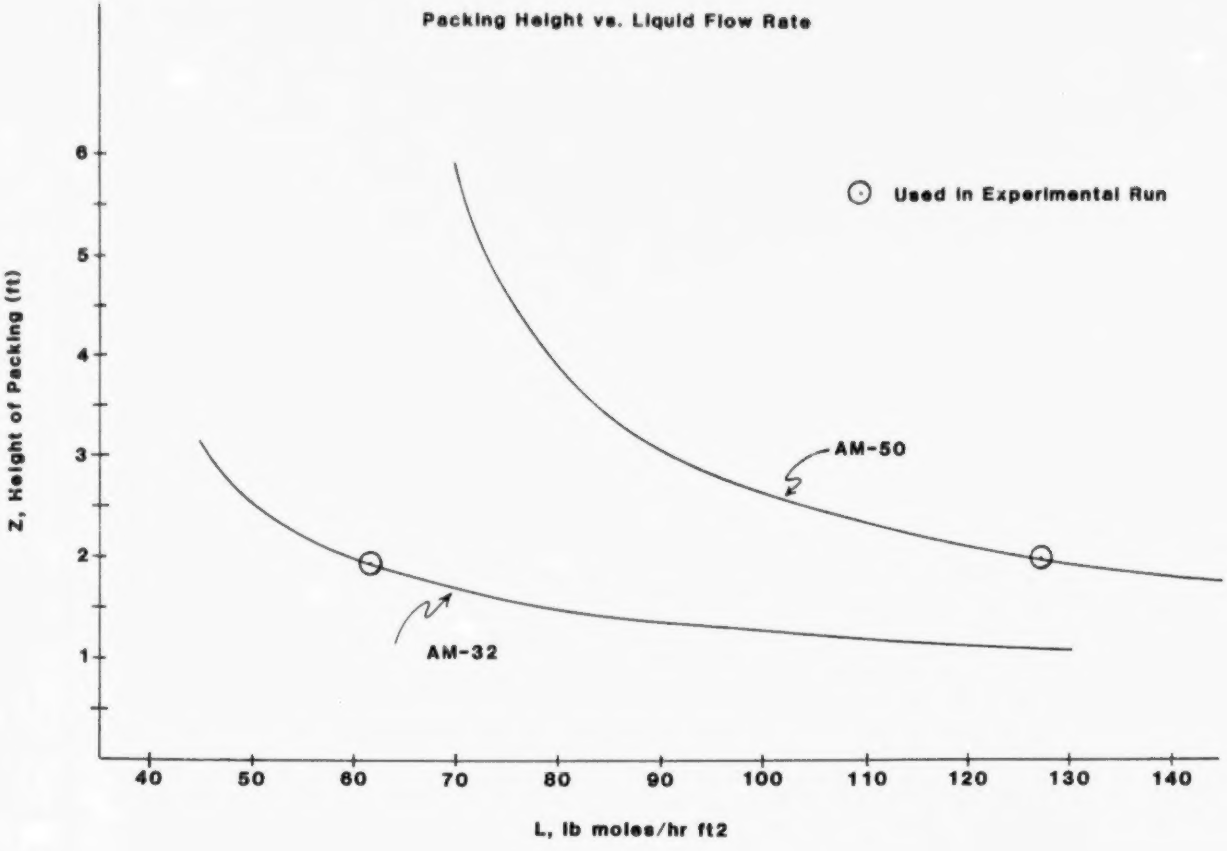


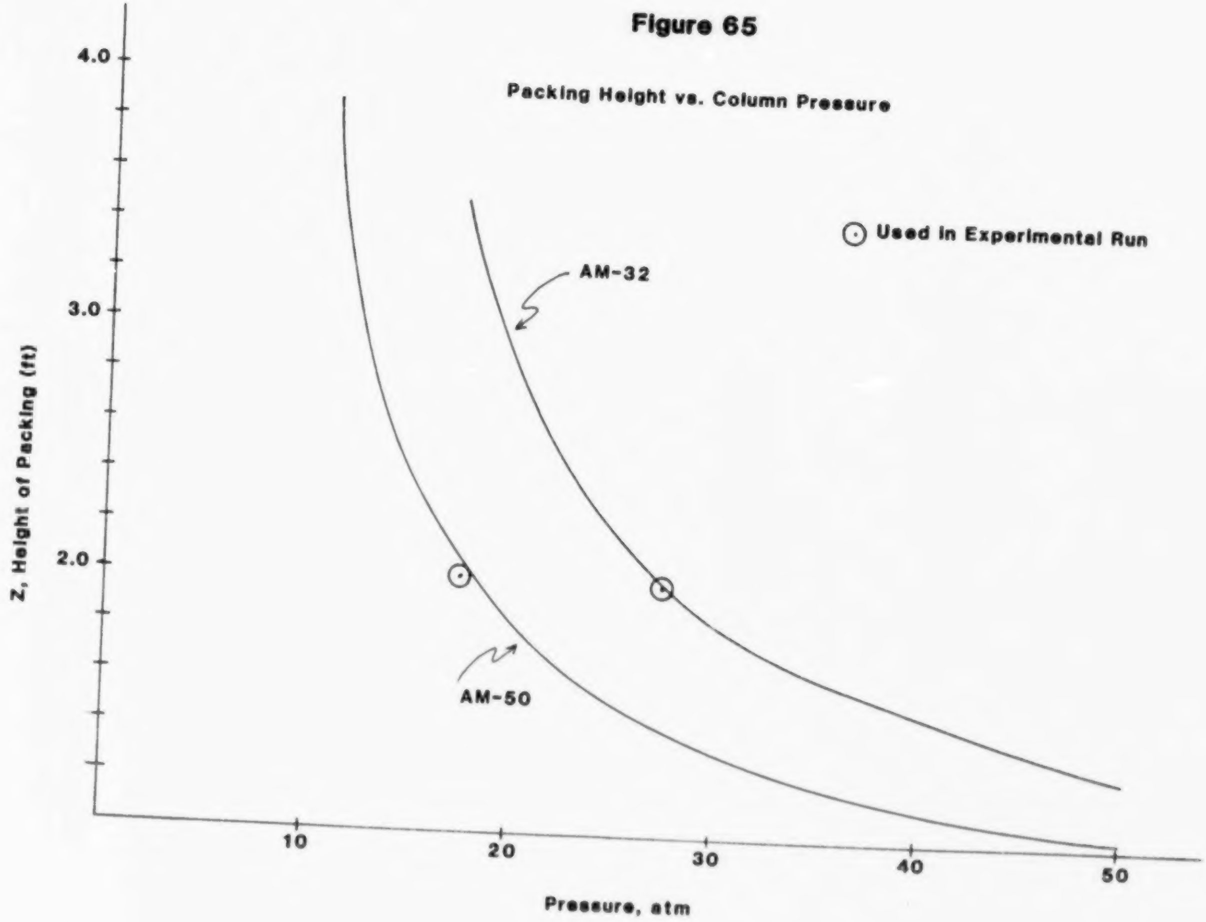
Figure 64



Packing Height vs. Liquid Flow Rate

Figure 65

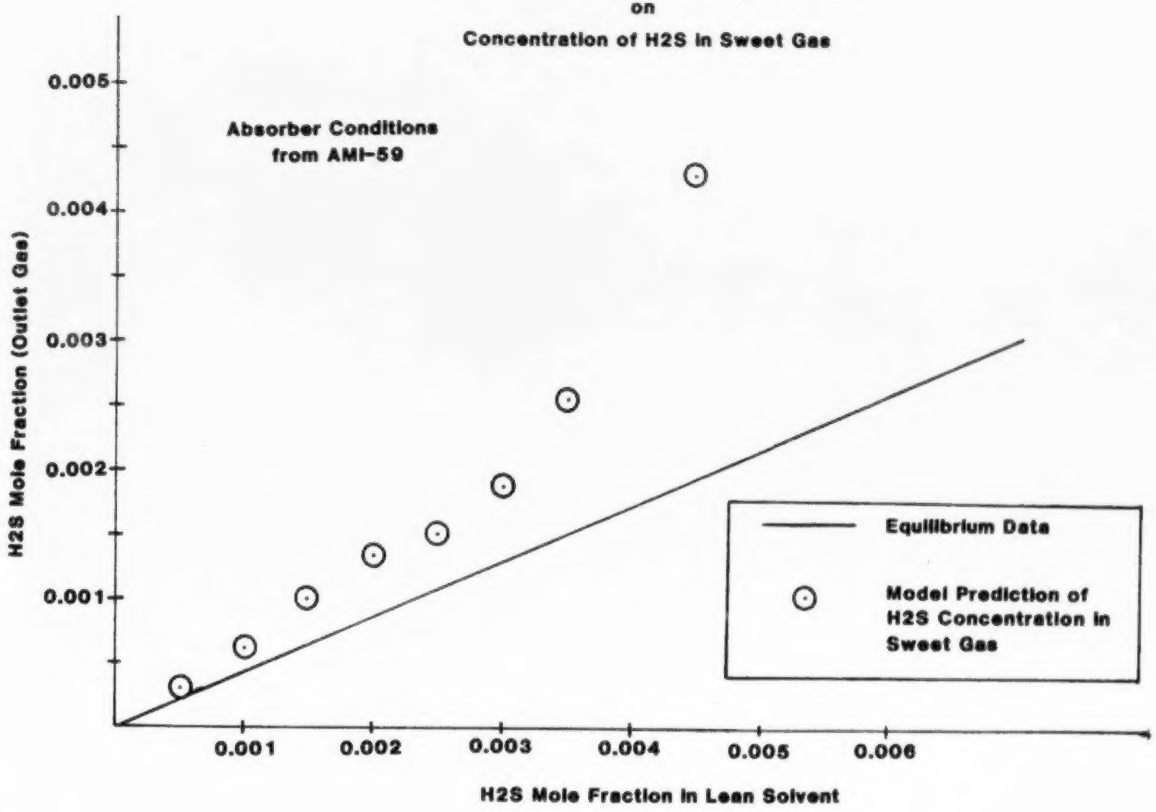
Packing Height vs. Column Pressure



content of the inlet liquid ranged from 0.0 to 0.0045 mole fraction. The effects of H_2S in the entering solvent on the outlet gas composition are presented in Figure 66, which shows that the H_2S in the exiting gas was nearly in equilibrium with the H_2S present in the entering solvent for the lower concentration ranges in Figure 66. At the higher concentrations of H_2S in the solvent, mass transfer considerations became more important as is evident from the difference from the equilibrium values. Hence, the removal efficiency for the entering H_2S in the gas stream was almost entirely a function of the amount of that compound in the entering solvent at liquid concentrations of less than 0.2 mole percent. This clearly illustrates the effect of stripper operation on absorber performance.

Figure 66

Effect of H2S Present in Inlet Solvent
on
Concentration of H2S in Sweet Gas



SUMMARY

The work described in this study formulates a mathematical model for the packed absorption column used in the work described by Ferrell et al (1982) in Volume 3 of this series of reports. This model, which is based on the algorithm developed by Feintuch and Treybal (1978, 1973) is essentially rigorous and has been shown to simulate absorber operation successfully over a wide range of operating conditions for both simple and multicomponent cases.

Additionally, the model has proved to be a useful way of determining the influence of a number of process variables on absorber performance. These analyses were performed on experiments involving carbon dioxide and hydrogen sulfide absorption by refrigerated methyl alcohol. Feed gases were prepared from gas cylinders or generated in a fluidized bed coal gasifier.

Future plans call for the extension of this model to describe packed stripping column performance and eventually to be part of a simulation package that will be used to describe the operation of the entire acid gas removal system.

LIST OF SYMBOLS

a, AA	interfacial area per unit volume of packing, area/ volume
$a_{dynamic}$	dynamic surface area per unit volume of packing, area/volume
a_e	effective surface area per unit volume of packing, area/volume
a_{max}	maximum effective surface area per unit volume of packing, area/volume
$a_{physical}$	effective surface area for physical absorption per unit volume of packing, area/volume
a_{static}	static surface area per unit volume of packing, area/volume
a_t, a_{tot}	total surface area per unit volume of packing, area/volume
a_{water}	effective area for aqueous systems per unit volume of packing, area/volume
a_w, a_{wet}	wetted surface area per unit volume of packing, area/volume
A	parameter used in Fair´s correlation
b	total liquid holdup in column, moles/area
b_j	holdup of j in column, moles/area
B	parameter used in Fair´s correlation
C_{F_L}	correction factor for mass transfer in liquid phase as a function of flooding velocity in Fair´s correlation
C_{G_j}	heat capacity of j in gas phase, energy/time-mass-degree

C_{L_j}	heat capacity of j in liquid phase, energy/time-mass-degree
C_G	heat capacity of gas phase, energy/time-area-degree
C_L	heat capacity of liquid phase, energy/time-mass-degree
d_c'	minimum of column diameter or 2, feet
d_p	nominal diameter of piece of packing, length
d_s	equivalent diameter of packing, length
D	diffusivity, area/time
EPS	total void fraction in packing, dimensionless
EPS_o	operating void space in packing, dimensionless
F_{G_j}	mass transfer coefficient of j in gas phase moles/time-interfacial area
F_{L_j}	mass transfer coefficient of j in liquid phase, moles/time-interfacial area
F_p	packing factor in Eckert's flooding velocity model
Fr	Froude number, $L'^2 a_t / \rho_L^2 g$
F_r	flooding ratio, dimensionless
f_μ	parameter used in Fair's correlation, dimensionless
f_ρ	parameter used in Fair's correlation, dimensionless
f_σ	parameter used in Fair's correlation, dimensionless
G_j	molar flow rate of j in gas phase, moles/time-area
G	gas molar flow rate, moles/time-area
G'	gas mass flow rate, mass/time-area

g	gravitational constant, length/time^2
$h_{dynamic}$	dynamic liquid holdup, volume per unit volume of packing
h_G	gas phase heat transfer coefficient, energy/time-area-degree
h_G'	gas phase heat transfer coefficient corrected for mass transfer, energy/time-area-degree
h_j	enthalpy per mole of gaseous component j, energy/mole j
h_L	liquid phase heat transfer coefficient, energy/time-area-degree
h_{static}	static liquid holdup, volume per unit volume of packing
h_{tot}	total liquid holdup, volume per unit volume of packing
$h_{w,tot}$	total liquid holdup for aqueous systems, volume per unit volume of packing
H_b	enthalpy of liquid holdup, energy
H_{ex}	enthalpy loss through column walls and other externals
H_G	enthalpy of the gas phase, energy
H_j	partial molar enthalpy of j in solution, energy/mole j
H_{ji}	partial molar enthalpy of j in solution at x_{ji} and t_i
H_L	enthalpy of liquid phase, energy
ΔH_{sj}	heat of solution of j, energy/mole j in solution
Henry_j	Henry's law constant, pressure
HTU_G	height of a gas phase transfer unit, length
HTU_L	height of a liquid phase transfer unit, length
k	Boltzmann's constant, dimensionless

k_G'	thermal conductivity of gas phase, energy/time-length-degree
k_L'	thermal conductivity of liquid phase, energy/time-length-degree
K_j	K-value of j, dimensionless
L_j	molar flow rate of j in liquid phase, moles/time-area
L	liquid molar flow rate, moles/time-area
L'	liquid mass flow rate, mass/time-area
m	constant used in Fair's correlation, dimensionless
M_A	molecular weight of A, mass/mole
M_B	molecular weight of B, mass/mole
MW_L	average molecular weight of liquid, mass/mole
n	constant used in Fair's correlation, dimensionless
N_j	mass transfer flux of j, moles/time-interfacial area
OMEGA	diffusion collision integral, dimensionless
p_j	partial pressure of j
P	pressure
Pr_G	Prandtl number of gas phase, $C_G*\mu_G/k_G'$
Pr_L	Prandtl number of liquid phase, $C_L*\mu_L/k_L'$
q_G	gas phase heat transfer flux, energy/time-area
q_L	liquid phase heat transfer flux, energy/time-area
Q_{ex}	external heat transfer flux, energy/time-area
R	gas constant
R_j	normalized molar flux of j, dimensionless
R_A	radius of a solute particle of A, length
Re_G	gas phase Reynolds number, dimensionless

Re_L	liquid phase Reynolds number, dimensionless
RATIO	ratio of Henry's law constant of key component to component j, at T and P
Sc_{Gj}	Schmidt number (gas) of j, $\mu_G/\rho_G{*}D_G$
Sc_{Lj}	Schmidt number (liquid) of j, $\mu_L/\rho_L{*}D_L$
SIGAB	characteristic length
SPEC	removal efficiency of key component, dimensionless
T	temperature
t_G	gas temperature
t_i	interfacial temperature
t_L	liquid temperature
t_o	reference temperature
v_A	atomic difussion volume of A
v_B	atomic difussion volume of B
V_A	liquid molar volume of A, volume/mole
V_B	liquid molar volume of B, volume/mole
V_j	partial molar volume of j, volume/mole
VMG	inlet molar gas flow rate, moles/time-area
VP	vapor pressure
We	Weber number, $L'^2/\rho_L \;\; a_t$
x_j	liquid mole fraction of j
x_{ji}	liquid mole fraction of j at interface
X_B	solvent association parameter
$XMOLG_j$	liquid mole fraction of j
y_j	gas mole fraction of j
y_{ji}	gas mole fraction of j at interface

z	height of packing, length
z_p	height of packing, length
λ_j	latent heat of vaporization, energy/mole
μ	viscosity, mass-length/time
σ	surface tension, force/length
σ_c	surface tension of packing surface, force/length
ρ	density, mass/volume
ψ	packing parameter in Fair´s correlation, length
ϕ	packing parameter in Fair´s correlation, length

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APPENDIX: NUMERICAL CONSIDERATIONS

In the process of developing a mathematical model for a packed absorber, provision must be made for mathematical discontinuities and convergence tolerances, as well as several other numerical considerations. Without proper handling, numerical factors can prevent any useful information from being obtained despite the appropriateness of the physical model.

In addition to mathematical difficulties, the convergence of several nested and overall iterative calculations depends upon the choice of a convergence tolerance. In other words, the difference between a trial and calculated variable must be smaller than a defined tolerance for the particular calculation to converge. In some cases, these tolerances are chosen in the program input while in other cases they are built into the program.

Finally, the time required for the convergence of a particular case is important from a practical standpoint. Excessively long convergence times diminishes the usefulness of a model for design or analysis. Some knowledge of the computation time required on a particular machine can be invaluable in choosing a design or analytical method.

Because of the importance of numerical considerations in any modeling effort, those that are important in MCOMP and SIMPAK will be discussed in this section.

MATHEMATICAL DISCONTINUITIES AND CONTINGENCIES

In both SIMPAK and MCOMP, there are several instances where a physically realistic situation can lead to a mathematical discontinuity. In the course of calculating iteratively a particular value, the calculation can be far enough, at times, from the true value that division by zero or the log of a negative number or zero may result. When these situations, which the computer has no way of handling, occur, the calculation may be abandoned unnecessarily. For this reason some contingency must be included in the program that will allow the calculation to continue without adversely affecting the final result. In both computer programs, checks are made prior to the use of any expression where a mathematical discontinuity could arise. If a problem exists, provision is made either to bypass the use of the particular expression or to assign temporarily a new value to the expression so that the calculation can continue. In this regard, many of the contingencies used by Feintuch (1973) and Khurana (1970) have been utilized and several others have been included to deal with these problems. In most instances where a problem occurs, a value computed in the previous iteration which has been stored is used so that the calculation can continue.

A contingency that has been developed for this application involves the molar flow gradients of the gas phase components in MCOMP. Several of the gaseous components are only slightly soluble in methanol for the range of the conditions studied here. For instance, in AMI-53, only 0.038 mole percent of H_2 is present in the exit liquid stream although the gas entering the column contained more than 30.0 mole percent H_2. As the calculation moves up the column, a situation can occur where the gradient used to calculate the amount of H_2 in the next packing segment in subroutine NSTEPM can be large enough to cause the mole fraction of H_2 in the methanol to become negative. This is, of course, physically unrealistic, and in such cases, a value of zero is assigned to the bulk liquid mole fraction of H_2. However, an additional problem arises because a very small amount of H_2 dissolved in the liquid gives rise to a substantial partial pressure; ignoring all H_2 in the liquid will make the calculated vapor phase composition meaningless. This problem becomes more serious when the presence of N_2, CO, and CH_4, all of which are only slightly soluble in methanol, are also considered.

The technique used to handle this problem was to set the bulk liquid mole fractions of any component equal to zero when a negative value was computed. In addition, the component is labeled and the most recently calculated interfacial liquid mole fraction is stored. In subsequent iterations, the value of the interfacial liquid mole fraction is used to compute the partial pressure of that component in the vapor phase. Although this technique introduces some inaccuracy, it allows the calculation to continue where otherwise it would generally fail.

In examining the output from MCOMP runs, the deviation of the calculated total pressure from the system pressure and the failure of the interfacial vapor phase mole fractions to sum to 1.0 are, in part, a result of using this contingency. Improvements to the handling of this problem are difficult to make because of possible round-off and truncation error problems that may arise from dealing with concentrations of less than 0.001 mole percent.

Most of the other contingencies used in both models are more straightforward and are evident in the computer code given by Kelly (1981) or have been discussed by Feintuch (1973) and Khurana (1970).

CONVERGENCE TOLERANCES

The basis for any iterative calculation is the comparison of a computed value to a previously assumed or trial value. Because these two values will rarely be equal, an acceptable tolerance must be established to decide when the iterations should end. While it would be desirable to make this tolerance as small as possible, computation time as well as round-off and truncation errors make this impractical. Instead the tolerance must be set so that there is a balance between the time required for convergence of the calculation and the accuracy of the result.

In the case of the computer programs used in this study, in which there are several nested iterative calculations, the problem of establishing tolerances for each calculation can become quite complex. In both SIMPAK and MCOMP, it is very important that the iterative calculations used in the subroutines TEMPI and MTEMPI converge in most instances. Recall that these subroutines coordinate three nested iterative calculations that are used to determine the values of the variables needed to compute the gas temperature gradient and the gas concentration gradient of the key component. These quantities are then used to compute the required information at the next level of packing. Failure of these calculations to converge necessitates use of the gradients calculated for the previous packing segment. This can lead to problems especially where the gradients are large. If this set of iterative calculations fails to converge for the first packing segment, the calculation is abandoned, often unnecessarily.

To minimize this problem in MCOMP, the convergence criteria are successively relaxed for the loop where the sum of the interfacial liquid mole fractions is compared to 1.0 (see Figure 8). After reading in an initial tolerance into the program, this value is increased by a total factor of 500 in five steps. For the computer runs made in this study, this range was usually 0.0001 to 0.05 mole fraction. While most of the time the most stringent criteria were met, situations where convergence was difficult were handled to allow the overall calculation to continue. This has been useful in dealing with situations where the normalized molar flux of the key component is very close to 1.0. A similar approach is used for this iteration in SIMPAK where the tolerance has been built into the model and is reduced from 0.0001 to 0.01 in two steps.

Within the loop that calculates the interfacial mole fractions, the interfacial gas mole fractions are calculated from both mass transfer and equilibrium relationships. In MCOMP, these two values must agree to within a tolerance read into the program as input. A value of 0.001% has generally been used. This value is relaxed to 10 times the original value if convergence is not achieved after 20 iterations. Built into SIMPAK for the same calculation are tolerances of 0.01% for the first 10 iterations after which the value is reduced to 1.0%.

In MCOMP, the convergence tolerances for the outlet gas flow rates are read into the model in the program input. It was found that agreement to within 1.5% or 0.005 lb-moles/hr-ft^2 yielded accurate results in a reasonable period of time. In SIMPAK, where only the outlet solvent flow rate need be determined iteratively, a tolerance of 1.0% has been built into the program.

In both programs, a trial and error calculation is used to determine the outlet liquid temperature (ENTHAL, ENTHM) and the temperature of the liquid in the next packing segment (NSTEP, NSTEPM). For a given liquid temperature, agreement between the calculated and assumed values of the energy balance must be within 0.1%. This has been built into both programs.

While there is probably room for improvement in both computation time and accuracy, the set of convergence tolerances used for this study were found to be adequate. For additional accuracy in calculations involving components at low inlet gas concentrations, like H_2S and COS, the tolerance of 0.005 lb-moles/hr-ft^2 on the outlet gas flows might be tightened but this will lead to longer computation times.

COMPUTATIONAL REQUIREMENTS

As mentioned previously, it is useful to know the computation time for the execution in a particular mathematical model on a particular machine. Recognizing this, the CPU (Central Processing Unit) time required for the convergence of MCOMP for AMI-36 was determined for three different machines commonly used for scientific computing purposes. The PDP 11/34 minicomputer with floating point hardware, the VAX 11/750 without floating point accelerator, and the VAX 11/780 with floating point accelerator; all of these systems are produced and marketed by the Digital Equipment Corporation of Maynard, Massachusetts, were used in this evaluation. The computation time requirements for the case of 50 packing increments were as follows:

MACHINE	CPU TIME (MINUTES)
PDP 11/34	88.47
VAX 11/750	23.57
VAX 11/780	6.92

This particular case required 9 overall iterations to converge which was the most required for any case submitted to MCOMP in this study. Usually less than 5 overall iterations were required for convergence so that this case represents a conservative estimate of the time required for model convergence for the conditions studied here.

Another important numerical factor is the chosen number of packing increments on the calculated height of packing. The calculated height of packing as a function of step size is shown in Figure 61 for the SIMPAK run for AM-32. Note that the calculated height of packing increases with the number of packing increments. This is because a larger number of packing increments results in a closer approximation to derivatives evaluated as required by the model.

It should be noted that for both SIMPAK and MCOMP the use of 50 packing increments provides a balance between accuracy and computation time. Since most of the work in this study was done on the PDP 11/34 minicomputer, only 10 packing increments were used in MCOMP to economize on computer time. This provided a reasonably accurate estimate of packing height and a relatively quick way to conduct a parametric study for several process variables in an optimization study. For final design estimates, the use of 50 packing increments is recommended.